



Določitev prispevka soljenja in posipanja cest na koncentracijo delcev PM₁₀



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1. Uvod

Na kakovost zraka v Sloveniji največ vplivajo emisije v zrak v sami državi, delno pa so tudi posledica transporta onesnaženosti zraka čez meje. Za pojavljanje povišanih koncentracij onesnaževal v zunanjem zraku so pomembni še drugi dejavniki, kot so klimatske značilnosti, meteorološki pojavi, fizikalno-kemijski procesi pretvorbe snovi v zraku in topografija. Aerosol je disperzni sistem, ki vsebuje tekoče in trdne delce suspendirane v plinu, ki ga imenujemo zrak. Delci so vpleteni v številne procese, sodelujejo pri različnih kemijskih in fizikalnih pretvorbah v onesnaženi atmosferi in pri nastanku kislega dežja vplivajo na vidnost in električne lastnosti atmosfere. Koncentracija in sestava delcev je odvisna predvsem od virov (naravnih in antropogenih) ter od meteoroloških pojavov. Različne študije so pokazale na povezavo med povišanimi koncentracijami delcev manjših od 10 μm in porastom boleznih respiratornega in kardiovaskularnega sistema.

Kjer so mejne vrednosti delcev PM_{10} v zraku presežene zaradi resuspenzije delcev po zimskem posipavanju ali soljenju cest, se takšno preseganje ne šteje za preseganje mejnih vrednosti po Uredbi o kakovosti zunanjega zraka (Ur.l.RS, št.9/2011).

Preseganja mejnih vrednosti za delce PM_{10} , pripisana zimskemu posipanju ali soljenju cest, se pri ocenjevanju skladnosti z mejnimi vrednostmi za kakovost zraka odštejejo, če so bili sprejeti ustrezni ukrepi za znižanje koncentracij PM_{10} . Pri obveščanju Komisije v skladu s členom 27, države članice predložijo potrebna dokazila, ki izkazujejo, da so vsa preseganja posledica takšne resuspenzije delcev in da so se sprejeli ustrezni ukrepi za znižanje koncentracij.

Prispevek zimskega soljenja smo določili na podlagi navodil s strani EK »Guidance on assessing the contribution of winter sanding and - salting under the EU Air Quality Directive«. Osnutka Smernice o zimskem posipanju in soljenju, ki temelji na EU Direktivi za kvaliteto zraka 2008/50/EC.

Agencija Republike Slovenije za okolje izvaja meritve delcev PM_{10} na trinajstih različnih merilnih mestih, ki delujejo v okviru državne merilne mreže. Od leta 2007 smo na različnih merilnih mestih opredelili vire delcev PM_{10} s pomočjo statističnega modela PCA. Iz omenjene analize virov smo ocenili, da je prispevek soljenja cest k celotni koncentraciji delcev PM_{10} okoli 10 %.

V letu 2010 smo zaradi vseh zgoraj navedenih dejstev odločili, da na enem merilnem mestu identificiramo in če je mogoče določimo prispevek zimskega soljenja na koncentracije PM_{10} . Odločili smo se za merilno mesto Maribor center, ker je to tipična prometna lokacija in ker je omenjeni prispevek v zimskem času največji. Merjenje PM_{10} se je izvajalo skozi leto 2010 (Januarja, Februarja, Marca, Oktobra, Novembra in Decembra).

2. Vzorčenje

Meritve delcev PM_{10} smo izvedli z merilnikom Leckel. To je avtomatski vzorčevalnik, z nizko volumskim pretokom. Vzorčevalnik je narejen za zunanje meritve pri vseh temperaturah in pogojih okolja. V magazinu je 15 filtrov, ki jih vzorčevalnik samodejno

menja ob datumu in uri, ki jo nastavimo. Časovna resolucija izpostavljenosti je bila 24 ur na enem filtru, pričetek je bil ob 0:00 po lokalnem času. Pretok zraka skozi vzorčevalnik je konstanten, in sicer $2,3 \text{ m}^3/\text{h}$. Kot medij za zbiranje delcev smo uporabili kvarčne filtre $\varnothing 47 \text{ mm}$, ki smo jih pred vzorčenjem žarili v žarilni peči 3 ure na $700 \text{ }^\circ\text{C}$.

3. Tehtanje

Ročno gravimetrično metodo izvajamo v skladu s standardom SIST EN12341:2000 in SIST EN 14907:2005. To pomeni delo v tehtalni sobi, v kateri so sledeči pogoji: relativna vlaga $50 \pm 5 \%$ in temperatura $20 \pm 1^\circ\text{C}$. Filtre kondicioniramo 48 ur pri zgoraj navedenih pogojih. Nato pričnemo s samim tehtanjem. Filtre stehtamo pred in po vzorčenju in iz razlike v masi ter iz pretoka zraka skozi filtre izračunamo koncentracijo PM_{10} delcev.

4. Kemijska analiza

Delci PM_{10} so bili analizirani na katione Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} in anione SO_4^{2-} , NO_3^- , Cl^- .

Za analizo ionov se uporabi 1/4 filtra. Filter se da v tubo kamor se doda 50 ml ultra čiste laboratorijske vode. Vse skupaj se ekstrahira 30 minut s pomočjo ultrazvočne kopeli. Po končani ekstrakciji je potrebno ekstrakt prefiltrirati ter ga nato odliti v dva viala (eden za določanje anionov in drugi za določanje kationov).

V ekstraktu filtra so anioni določeni z ionskim kromatografom Dionex DX – 120, na koloni IonPack AS14, dimenzije $4 \times 250 \text{ mm}$. Na kolono se injicira $100 \mu\text{L}$ vzorca. Predkolona služi za zaščito kolone, medtem ko supresor zmanjša prevodnost ozadja, kar olajša detekcijo. Katione pa se v ekstraktu filtra določi tudi s pomočjo ionskega kromatografa, in sicer z Waters 431, na koloni IC-Pak Cation M/D, dimenzije $3,9 \times 150 \text{ mm}$, z inštalirano predkolono. Na kolono se prav tako injicira $100 \mu\text{L}$ vzorca. V obeh inštrumentih je za detekcijo ionov uporabljen detektor za merjenje prevodnosti.

5. Merilno mesto Maribor

To merilno mesto ima tipične karakteristike »cestnega kanion-a«. 50 m stran se nahaja križišče s svetlobno signalizacijo. Merilno mesto se nahaja tik ob Titovi cesti. Ta lokacija je ena najbolj onesnaženih lokacij v Sloveniji, katerih glavni onesnaževalec je promet, saj se tukaj dnevno pelje kar 46 000 avtomobilov.

6. Rezultati

Merilno mesto Maribor ima tipične karakteristike urbane lokacije z visokim pretokom prometa. V našem projektu se je merjenje PM_{10} izvajalo skozi leto 2010 (januarja, februarja, marca, oktobra, novembra in decembra). Obdelali smo vse vzorce z PM_{10} , ki so imeli koncentracijo višjo od $40 \mu\text{g}/\text{m}^3$.

Iz rezultatov je razvidno, da je koncentracija $50 \mu\text{g}/\text{m}^3$ (mejna dnevna vrednost) prekoračena 47 krat v času od januarja do decembra 2010. Koncentracija PM_{10} je bila višja kot $100 \mu\text{g}/\text{m}^3$ 6 krat (18.1., 28.1, 17.2., 18.2., 30.12. in 31.12.). Najvišja izmerjena koncentracija PM_{10} je bila $127 \mu\text{g}/\text{m}^3$, in sicer 18.2.2010. Koncentracija klorida in natrija ni neposredno odvisna od koncentracije PM_{10} . Ponavadi je razmerje med kloridom in natrijem 2:1 in 1:1. Razmerje 2:1 je pričakovano zaradi vira soljenja. Večja koncentracija NO_3^- , kaže tudi na onesnaženje iz antropogenih virov (velika verjetnost prometa). Največja koncentracija NO_3^- je bila $20 \mu\text{g}/\text{m}^3$ 12.1.2010. Najmanjša koncentracija klorida je bila izmerjena 11.10.2010 in je znašala $0,11 \mu\text{g}/\text{m}^3$, največja pa je bila izmerjena 28.1.2010 in je znašala $6,89 \mu\text{g}/\text{m}^3$. Najnižja zaznana koncentracija natrija je bila $0,05 \mu\text{g}/\text{m}^3$ 8.10.2010 in najvišja $3,99 \mu\text{g}/\text{m}^3$ 27.1.2010.

Prevladujoči anioni so bili NO_3^- in SO_4^{2-} . Med kationi prevladuje NH_4^+ . Koncentracije Ca^{2+} , Mg^{2+} in Na^+ , so bile zelo podobne. Za vse merilne mesece v letu 2010 prevladujejo sledeče komponente PM_{10} : NO_3^- , SO_4^{2-} in NH_4^+ . Če primerjamo zimске in jesenske mesece ugotovimo, da je v zimskem času izmerjena koncentracija Cl^- približno 40 % večja. Prav tako smo ugotovili, da je povišana koncentracija Na^+ (26 %).

V jesensko zimskih mesecih (koncentracije nad $40 \mu\text{g}/\text{m}^3$) se koncentracije Cl^- gibljejo med $0,19$ do $1,32 \mu\text{g}/\text{m}^3$. Opazne so razlike v dnevih, ko so bile ceste soljene in ko soljenje ni bilo prisotno. Zlasti januarja so bile koncentracije za 30 % višje v soljenih dneh ko ni bilo prisotno soljenje. Vpliv soljenja je viden po približno treh dneh po soljenju.

Razlika v koncentracijah anionov in kationov med meseci, ko je bilo prisotno soljenje (januarja, februarja, marca in decembra 2010) in brez soljenja (oktober, november 2010) je jasno razvidna. Primerjava med zimskim in jesenskim obdobjem, kaže, približno 4-krat višje vrednosti za Cl^- v zimskih mesecih.

Za obdobje od januarja do marca je povprečna koncentracija klorida 1,4 % celotne koncentracije PM_{10} za vse dni s soljenjem in dneve brez soljenja. Za oktober in november 2010 znaša koncentracija Cl^- približno 0,6 % celotne koncentracije PM_{10} . Avstrijski model pa kaže, da je bila povprečna koncentracija klorida v PM_{10} za 4 % večja v dneh, ko je bilo prisotno zimsko soljenje, in 3 % v dneh brez soljenja.

7. Veter

Na merilni lokaciji v Mariboru je veter usmerjen severno-zahodno in ima zelo majhno hitrost ($0-4 \text{ m}/\text{s}^2$). Iz teh podatkov o majhni hitrosti vetra lahko zaključimo, da je daljinski prispevek h koncentraciji Cl^- zelo malo verjeten (soljenje iz drugih ulic).

8. Zaključki

Prispevek soljenja k povišanim koncentracijam PM_{10} pozimi je prav gotovo razviden:

- ne takoj, vendar pogosto po soljenju (2-3 dni).
 - glavni meteorološki pogoji, ki povečujejo koncentracijo PM_{10} in Cl^- so: suho vreme, nizka relativna vlažnost zraka in nizka hitrost vetra.
- Vpliv soljenja je razviden iz:
- običajno večje koncentracije PM_{10} ,
 - višje koncentracije Na^+ in Cl^- ,
 - razmerja med Na^+ in Cl^- , ki je med 1:1 in 1:3.3,
 - najvišje izmerjena koncentracija Cl^- , ki je znašala $6,9 \mu g/m^3$.

Iz rezultatov analiz smo izračunali, da je prispevek soljenja cest k celotni koncentracij delcev PM_{10} le $2 \mu g/m^3$. Število preseganj na letni ravni ostane isto.

Kontaktirali smo tudi institucije v Avstriji in dobili sledeč odgovor: da je delež klorida v delcih PM_{10} zaradi soljenja cest prenizek in da s tem število preseganj delcev PM_{10} ne zmanjša v takšni meri, da bi zadostili zahtevam 14. člena Uredbe o kakovosti zunanjega zraka.

9. Priloga

Poročilo: QUANTIFICATION OF THE CONTRIBUTION OF WINTER SALTING TO PM_{10} IN MARIBOR

PRILOGA 1

UNIVERSITY OF NOVA GORICA
SCHOOL OF ENVIRONMENTAL SCIENCES

**QUANTIFICATION OF THE CONTRIBUTION OF
WINTER SALTING TO PM₁₀ IN MARIBOR**

PROJECT I

Leonida Mehle

Mentor: doc. dr. Irena Grgić

Nova Gorica, Februar 2011

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1. INTRODUCTION

Since the industrial revolution the quality of air has deteriorated considerably - mainly as a result of human activities. Increase in the industrial and energy production, a high consumption of fossil fuels and the dramatic increase in traffic, etc. all contribute to high air pollution in our environment which, in turn, can lead to serious health problems. For example, air pollution is increasingly being cited as the main cause of lung affection such as asthma; twice as many people suffer from asthma today compared to 20 years ago.^{13,14}

Nowadays air quality is now a major concern for many European citizens. It is also one of the areas in which the European Union has been the most active. Since the early 1970s, the EU has been working on improvement of air quality by controlling emissions of harmful substances into the atmosphere, improving fuel quality and by integrating environmental protection requirements into the transport and energy sectors.^{1,2} In developed countries, two thirds of the population lives in urban areas comprising of only about one percent of the land. The growth of suburban, and new superhighways have made it possible for more people to travel greater distances and thus to converge faster to our cities. Hence, an increasing population combined with a high standard of living has led to drastically intensified and concentrated air pollution in localized areas.

Aerosol particles are ubiquitous in earth's atmosphere and are vital to many environmental issues such as climate change and air quality.^{6,7} They also represent an important risk factor for various adverse health effects in humans. The concentration and size distribution of atmospheric aerosol particles are highly variable in space and time.^{9,10,11,12} In the last few years a spectacular growth of interest and progress in the field of aerosol science can be noticed, particularly on the properties of aerosols, their sources, processes and their environmental impact.

It is well known that in the areas with a cold winter climate special air pollution problems can occur. For example, particulate matter (PM) can accumulate in the urban environment over the winter months and deposit on the ground during snowing. Particles can then be released into the atmosphere. In the urban environment the PM₁₀ levels can rise drastically, especially in the proximity of heavy traffic roads when their surfaces dry out.^{1,2} Special meteorological conditions, such as ground based low-height inversions with low wind speeds, are often related to the high PM concentrations. In addition, winter-sanding and -salting may also contribute to higher PM concentrations.

Another interest towards the composition and sources of the springtime dust rises from the new EU limit values for airborne particles. If the EU limit values given in the Council Directive 1999/30/EC for thoracic particles (PM₁₀) are exceeded, member states must implement action plans in accordance with Council Directive 1996/62/EC for attaining the limit value within a specific time limit. However, the member states can designate the zones within which the limit values are exceeded due to the resuspension of particulates following the winter sanding of roads (Council Directive 1999/30/EC, article 5). A list of such areas must be provided with information of concentrations and sources of PM₁₀. It must be shown that the exceedances are due to road sanding and that reasonable measures have been taken to lower the concentrations.

Directive 2008/50/EC on ambient air quality and cleaner air for Europe must be transmitting in the legislation of the Republic of Slovenia.^{3,5} On the basis of that directive the guidance is made. Guidance consolidates the majority of existing legislation on air pollution and further

specifies maximum levels of contaminants. Directive 2008/50/EC give the 24-hour annual values for PM₁₀ particles. For Slovenia, it is reported that winter sanding and salting should be measured and evaluated according to Directive 2008/50/EC.

The aim of the project is to identify, and if possible to quantify, the impact of winter-salting on the PM₁₀ concentration on the basis of the guidance on the quantification of the contribution of natural sources under the EU Air Quality Directive 2008/50/EC. In the project the data for PM₁₀ concentrations for the winter months from one selected sampling site Maribor, Environmental Agency of the Republic of Slovenia were collected. The aims of this project can be divided into the following main points:

- To identify and if possible quantify the impact of winter-salting on PM₁₀ concentrations.
- To recommend the methods for determination of contributions of winter-salting to PM₁₀ exceedances (based on the experience from Finland and/or Austria).

For the purpose of this project the following data were also obtained:

- Meteorological conditions.
- Information on the days when roads have been salted.

This project should demonstrate that winter-salting is the actual and only source of the contribution to the elevated PM₁₀ concentrations. Contributions from other PM₁₀ sources that might have a similar influence on PM₁₀ levels should be excluded. The quantification of the winter-salting must be as precise as possible. With respect to air quality plans dealing with PM₁₀, the results apply to the representative area of the affected monitoring site.

2. THEORETICAL PART

2.1 Definitions

The impact of winter-salting is analysed on a daily basis. Winter-sanding-salting influences the PM₁₀ concentrations on a time scale of some hours or some days and has to be investigated on at least a daily basis.⁵ This is also the time scale for the more relevant PM₁₀ limit value, which is more stringent than the annual average limit value. The influence of winter-salting on the annual mean concentration is very difficult to determine with sufficient accuracy without analysing single days. Compliance with the annual limit value may be checked by calculating the annual mean value from the daily mean values reduced by the contribution from winter-sanding and -salting.

Member States may under the EU Air Quality Directive 2008/50/EC designate zones or agglomerations within which limit values for PM₁₀ are exceeded in ambient air due to the re-suspension of particulates following winter-salting of roads. Guidance on assessing the contribution of winter-salting under the EU Air Quality Directive recommends methods for the determination of the contributions of these processes to the concentrations.

These contributions have to be determined on a daily basis and can then be subtracted from each measured daily mean PM₁₀ value. When also the annual mean limit value is exceeded, a reduced annual mean value can be calculated using the reduced daily mean values.

2.2 Identification of the influence of winter-salting on PM₁₀ levels

For the purpose of subtracting contributions to PM₁₀ limit value exceedances attributable to winter-salting it is necessary to provide a robust demonstration and quantification of the actual contribution from these sources to the concentrations:

- Contributions must be attributed unequivocally to winter-salting activities,
- Quantification must be sufficiently precise and accurate and appropriate to the averaging period of the limit value,
- Representativeness of the affected monitoring station,
- The contribution of winter-salting to the measured PM₁₀ concentration has to be quantified in µg/m³ for each exceedance day,
- Chemical analysis of chloride for winter-salting,
- The method to determine this contribution has to be documented.

2.2.1. Contributions must be attributed unequivocally to winter-salting activities

It is needed to be demonstrated that winter-salting is the actual and only source of contribution to the PM₁₀ concentrations regarded as such.

2.2.2. Quantification must be sufficiently precise and accurate and appropriate to the averaging period of the limit value

The quantification of the winter-salting must be as precise as possible. The quantification of the contribution from winter-salting has to be performed, as a first step, for candidate days exceeding the concentration of 50µg/m³.

2.2.3. Representativeness of the affected monitoring station

Increased PM₁₀ emissions by winter-salting usually affect the PM₁₀ concentration measured at traffic related monitoring sites. The procedure to subtract the contribution of winter-salting is to be applied for each monitoring station separately. With respect to air quality plans dealing with PM₁₀, the results apply to the representative area of the affected monitoring site.

2.2.4. The contribution of winter-salting to the measured PM₁₀ concentration has to be quantified in µg/m³ for each exceedance day

For each day for which a reduction due to winter-salting is applied, this reduction has to be quantified as a concentration. It is not sufficient to “remove” the daily PM₁₀ value.

2.2.5. Chemical analysis of chloride for winter-salting

Winter-salting means dispersing salt (NaCl, CaCl₂, MgCl₂, calcium-magnesium-acetate, sulfates, nitrates, ammonium, etc.) or a brine or solution of a salt for traction control on the road surface or pavement.

In most non-coastal areas of Europe, winter-salting is the only source of chloride. The total salt concentration can – based on the chemical composition of the salt used – be determined by the chloride concentration in PM₁₀. Exceedances due to the effect of winter-salting may be shown for exceedance days using chemical analysis of 24-hour PM samples.

This method requires daily chemical analyses of the relevant PM₁₀ component of those days for which a winter-sanding or -salting subtraction is intended to be applied. In case of salting, this is in most cases a substance containing chloride. The corresponding information on the chemical properties of the salt dispersed on the road is necessary.

The chemical analysis of chloride (or other components of salt dispersed on the road, if it is not a chloride salt) is assumed to be a well suited method to quantify the impact of winter-salting on PM₁₀ concentrations.

2.2.6. The method to determine this contribution has to be documented

The method has to be documented as a reference to a report or a publication, which gives information about the input data used and the assumptions on which estimates are based.

2.3 Procedures to assess the impact of winter-sanding and –salting

Increased PM₁₀ concentrations due to “winter related road traffic emissions” are recognised as a problem in several northern regions, both in Europe and North America. The most thorough research related PM₁₀ problems seems to have been conducted in Finland.²⁹ Austrian data on the chemical speciation of PM₁₀ have been analysed in more depth.¹⁷

2.3.1. Austrian data

The availability of different types of data and information from Austria facilitated the evaluation of these data for the possible identification of an impact of winter-salting. Besides the PM₁₀ values themselves, data on the chemical composition and the size distribution were used.

Most data on the chemical composition have been taken from a project entitled “AQUELLA”;^{25,26,27,28} in this project; chemical analyses were averaged over several days.

In comparison to northern Europe, the PM_{2.5}/PM₁₀ ratios are quite high, and the difference between PM₁₀ and PM_{2.5} – in absolute concentrations – is low. The number of exceedances is being reduced by only a minor amount. The criterion used in Finland (comparison the PM_{2.5}/PM₁₀ ratio, difference between PM₁₀ and PM_{2.5}, etc.) for long-range transport is not applicable in Austria, since situations with long-range transport are usually associated with high rural background PM_{2.5} concentrations.

To conclude, in central European countries such as Austria, the relation of PM_{2.5} to PM₁₀ does not exhibit a clear pattern related to winter-sanding (or -salting). Significant differences compared to northern Europe concern:

- High rural background PM_{2.5} concentrations.
- Low urban increments both for PM₁₀ and PM_{2.5}.
- Low coarse fractions even at kerbside locations.

Data on chemical composition of PM₁₀ from Austrian monitoring sites were analysed with respect to a possible influence of winter-sanding.^{25,26,27,28} Indicators for winter-salting are the fractions of mineral material and chloride in PM₁₀. The only source of chloride in Austria is winter-salting, since sea spray is of no relevance. Mineral material, however, may originate from various sources. Besides winter-sanding, the re-suspension of any dust by road traffic as well as road abrasion, industrial (fugitive) sources, erosion from agricultural or barren

surfaces or desert dust may cause elevated mineral material concentrations. They found that chloride concentrations are about twice as high on days with winter-salting; the difference to days without salting is less for days with PM₁₀ concentrations above 50 µg/m³.

3. PURPOSE OF WORK

The main purpose of the project was to estimate the contribution from winter-salting to PM₁₀ concentrations in the area of Maribor on the basis of measurements with the reference sampler Leckel. Chemical analysis of PM₁₀ samples were performed in the Chemical analysis laboratory of the Environmental Agency of the Republic of Slovenia (ARSO). Anions were determined according to the standard EN 10304-1, cations were determined according to the standard EN 14911.

Winter salting of roads can contribute significantly to an excess of limit values for PM₁₀ (50 µg/m³). Where it is possible to measure this contribution, the EU Air Quality Directive allows Member States to deduct this contribution from the total concentration to demonstrate compliance.^{16,17,18} It is necessary to show that the winter salt is the only real source of the contribution to the elevated PM₁₀ concentrations. In our work the procedure proposed by Austrian model^{25,26,27,28} has been considered.

4. SAMPLING AND MEASUREMENT METHOD

4.1 Measuring site Maribor

This measuring site has a typical character of a street canyon. The measuring site is located on a main road leading to the center of town. 50 m away is a crossing with traffic lights. Nearby is a bus station. The container after Tito's road is located at the measuring point. The chosen location is one of the more polluted in Slovenia due to heavy traffic - about 46000 vehicles per day.



Figure 1: Measuring site Maribor¹⁹

4.2 PM₁₀ measurements

Measurements of PM₁₀ particles were performed during the year 2010 (January, February, March, October and November) with a reference sampler Leckel. This is an automatic low volume sampler (LVS), which is designed for field measurements at all temperatures and environmental conditions. Fifteen filters in a set are changed automatically at defined times.^{8,20,21,22,23}

The air flow through the sampler is 2.3 m³/h. The entire sampling system is cooled by air flow. The minimum sampling time of one filter is 1 h, the maximum time is 168 h. The sampler was placed on the roof of the container, at a height of 3.5 m. The sampling time was 24 h, the start being at 0:00 h after local time. PM particles were collected on Whatman quartz filters, which were weighed by using a Sartorius balance. Filters were heated at 500° C for 3 h before sampling.

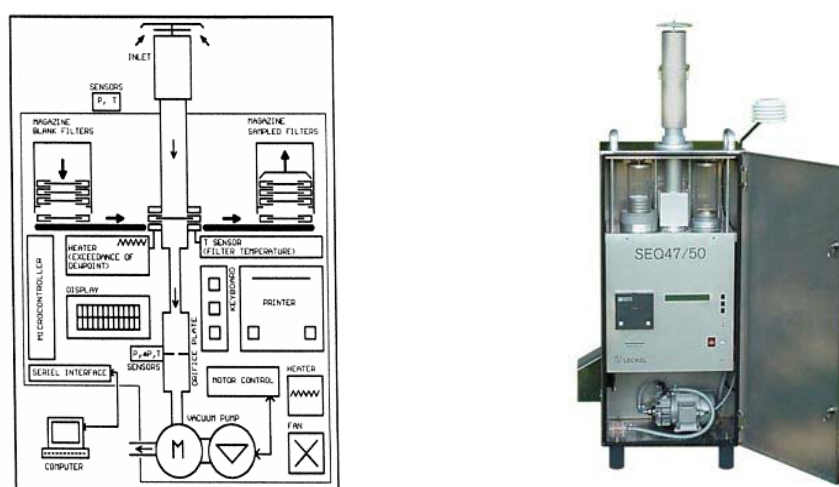


Figure 2: Schematic scheme for particulate sampling²⁴

The weighing of the filters was performed according to standards EN 12341: 2000 and EN 14907: 2005. Filters were weighed before sampling and after sampling, after conditioning for 48 hours at a relative humidity of 50 ± 5 % and temperature 20 ± 1 °C.

4.3 Determination of anions and cations

A part of the filter was extracted for 30 min in 10 mL Milli-Q water in an ultrasonic bath. The extracts were filtered through a syringe filter (pore size 0.2 µm). The content of anions (Cl⁻, NO₃⁻ and SO₄²⁻) and cations (Na⁺, K⁺, NH₄⁺, Ca²⁺ and Mg²⁺) was determined by ion-exchange chromatography. The identification of each ion is based on the retention time, while the concentration was determined using a calibration curve based on the peak area. Determination of anions was carried out with a separation column Dionex Ion Pac AS14, and the background was reduced by a suppressor (Dionex ASRS Ultra II). A mixture of 3.5 mM Na₂CO₃ and 1.0 mM NaHCO₃ was used as an eluent. For the detection of chloride and sulfate a conductivity detector was used. Nitrate was detected spectrophotometrically at a wavelength of 210 nm. The cations separation was carried out using Waters IC PAK column Cation. A mixture of 0.1 mM EDTA and 3.0 mM HNO₃ was used as an eluent. Cations were detected by means of a conductivity detector.

5. RESULTS AND DISCUSSION

The measuring site in Maribor is a typical urban location with heavy traffic. Using statistical models, ARSO demonstrated that the impact of the salting in Maribor in 2007 was 11 %.¹⁸ However, due to limited financial resources they have a limited number of measurements, so the results are uncertain. In our project, measurements have taken place during the year 2010 (January-March, October and November).

In Tables 1-5 the results for mass concentrations and concentrations of anions and cations in PM₁₀ particles collected in January, February, March, October and November are presented. However, only the samples with the PM₁₀ concentration higher than 40 µg/m³ were taken into account.

Table 1: Mass concentrations and concentrations of anions and cations in PM₁₀ samples from Maribor, January 2010

Date	PM ₁₀	NO ₃ ⁻	NH ₄ ⁺	K ⁺	SO ₄ ²⁻	Ca ²⁺	Cl ⁻	Na ⁺	Mg ²⁺
	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³
5.1.2010	46	9.3	3.1	0.53	2.54	0.31	0.51	0.38	0.06
6.1.2010	60	10.3	6.0	0.71	7.26	0.18	1.11	0.73	0.05
7.1.2010	63	11.8	6.5	0.62	8.89	0.16	0.54	0.34	0.04
8.1.2010	42	8.2	4.2	0.53	4.72	0.13	0.40	0.25	0.03
11.1.2010	48	9.4	4.5	0.53	5.26	0.14	0.44	0.25	0.04
12.1.2010	75	20.0	8.9	0.71	8.34	0.17	0.38	0.25	0.05
13.1.2010	53	9.8	4.9	0.47	6.17	0.22	0.34	0.17	0.06
14.1.2010	63	11.6	7.1	0.67	10.16	0.20	0.44	0.12	0.06
15.1.2010	64	11.4	6.5	0.60	8.35	0.24	0.31	0.18	0.06
16.1.2010	51	4.2	3.8	0.44	8.71	0.25	0.33	0.24	0.31
17.1.2010	70	9.1	6.2	0.62	9.43	0.12	0.38	0.27	0.11
18.1.2010	102	15.5	9.5	1.13	14.06	0.29	0.91	0.31	0.10
19.1.2010	74	14.5	6.3	1.03	6.17	0.36	0.38	0.56	0.14
20.1.2010	41	8.0	2.5	0.54	2.54	0.44	1.43	1.00	0.13
21.1.2010	51	8.9	4.3	0.44	6.07	0.27	0.70	0.49	0.07
22.1.2010	66	12.3	6.2	0.58	8.71	0.20	0.91	0.67	0.04
23.1.2010	80	13.8	8.5	0.80	13.97	0.24	0.34	0.53	0.06
24.1.2010	89	16.0	9.3	1.05	14.33	0.15	0.18	0.31	0.04
25.1.2010	69	7.3	7.3	0.78	15.23	0.25	0.71	0.54	0.08
26.1.2010	75	8.0	6.2	0.71	11.61	0.25	0.45	0.36	0.07
27.1.2010	69	6.9	4.0	0.60	6.53	0.44	3.27	1.81	0.13
28.1.2010	119	15.1	4.7	1.05	7.62	1.05	6.89	3.99	0.27
29.1.2010	79	10.5	4.2	0.74	5.08	0.56	2.00	1.20	0.14

Table 2: Mass concentrations and concentrations of anions and cations in PM₁₀ samples from Maribor, February 2010

Date	PM ₁₀	NO ₃ ⁻	NH ₄ ⁺	K ⁺	SO ₄ ²⁻	Ca ²⁺	Cl ⁻	Na ⁺	Mg ²⁺
	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³
2.2.2010	59	5.6	1.2	0.47	1.6	0.85	4.35	2.36	0.42
3.2.2010	63	5.6	1.3	0.47	1.4	1.02	4.17	2.72	0.24
4.2.2010	45	6.9	2.2	0.40	1.6	0.49	1.36	1.02	0.14
5.2.2010	50	8.5	3.1	0.54	2.7	0.40	0.49	0.36	0.08
6.2.2010	42	7.3	4.7	0.54	6.5	0.13	0.29	0.24	0.03
7.2.2010	45	4.7	4.7	0.45	3.1	0.15	0.22	0.38	0.06
8.2.2010	56	6.0	6.3	0.54	11.8	0.16	0.20	0.27	0.04
9.2.2010	65	7.8	7.4	0.60	14.5	0.22	0.56	0.24	0.05
10.2.2010	67	7.1	7.5	0.69	13.1	0.21	0.21	0.26	0.05
11.2.2010	40	4.5	4.2	0.51	8.2	0.17	0.22	0.31	0.05
12.2.2010	78	6.9	5.1	0.51	9.4	0.33	0.33	0.27	0.16
13.2.2010	54	11.6	8.3	0.73	12.2	0.22	0.69	0.62	0.06
14.2.2010	52	7.4	5.4	0.56	9.6	0.45	0.63	0.54	0.10
15.2.2010	62	6.5	4.7	0.49	8.7	0.63	0.49	0.33	0.22
16.2.2010	76	11.8	6.5	0.69	9.8	0.47	0.74	0.56	0.14
17.2.2010	119	18.1	10.7	1.1	13.2	0.44	0.85	0.60	0.11
18.2.2010	127	23.6	12.7	1.3	14.0	0.60	0.67	0.42	0.13
19.2.2010	61	10.3	4.5	0.6	4.9	0.63	0.85	0.49	0.15
23.2.2010	52	1.7	0.20	0.22	0.82	1.05	0.87	0.42	0.58

Table 3: Mass concentrations and concentrations of anions and cations in PM₁₀ samples from Maribor, March 2010

Date	PM ₁₀	NO ₃ ⁻	NH ₄ ⁺	K ⁺	SO ₄ ²⁻	Ca ²⁺	Cl ⁻	Na ⁺	Mg ²⁺
	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³
12.3.2010	50	6.5	4.72	0.53	7.07	0.54	1.00	0.51	0.08
17.3.2010	44	5.8	2.00	0.24	2.18	1.07	1.43	1.00	0.17
18.3.2010	58	7.4	2.00	0.36	2.18	1.47	1.18	1.02	0.25
19.3.2010	60	11.1	3.63	0.40	3.45	0.22	1.14	1.03	0.29
20.3.2010	44	8.9	3.81	0.29	4.35	0.78	0.51	0.33	0.20
23.3.2010	42	2.9	1.02	0.20	1.22	0.60	0.96	0.71	0.14
24.3.2010	46	3.1	1.23	0.20	1.81	0.62	0.54	0.44	0.15
25.3.2010	58	4.2	3.63	0.36	8.71	0.98	0.54	0.49	0.20
26.3.2010	46	1.8	1.34	0.17	3.45	0.93	0.44	0.45	0.13

Table 4: Mass concentrations and concentrations of anions and cations in PM₁₀ samples from Maribor, October 2010

Date	PM ₁₀	NO ₃ ⁻	NH ₄ ⁺	K ⁺	SO ₄ ²⁻	Ca ²⁺	Cl ⁻	Na ⁺	Mg ²⁺
	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³
8.10.2010	43	1.8	1.5	0.22	4.2	0.82	0.12	0.05	0.14
9.10.2010	44	3.4	2.0	0.33	5.1	1.1	0.13	0.06	0.18
11.10.2010	44	2.4	1.2	0.27	2.5	0.91	0.11	0.06	0.14
12.10.2010	48	4.2	1.7	0.34	3.1	0.91	0.18	0.07	0.17
13.10.2010	49	4.2	2.0	0.42	3.4	0.74	0.18	0.06	0.17
14.10.2010	60	8.0	3.4	0.47	5.4	0.93	0.16	0.08	0.20
15.10.2010	74	12.2	4.7	0.51	6.3	0.91	0.31	0.08	0.20
16.10.2010	46	6.3	2.9	0.33	4.2	0.51	0.20	0.07	0.15
23.10.2010	46	4.4	1.2	0.45	1.7	0.73	0.33	0.17	0.16

Table 5: Mass concentrations and concentrations of anions and cations in PM₁₀ samples from Maribor, November 2010

Date	PM ₁₀	NO ₃ ⁻	NH ₄ ⁺	K ⁺	SO ₄ ²⁻	Ca ²⁺	Cl ⁻	Na ⁺	Mg ²⁺
	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³	µg/m ³
2.11.2010	48	2.9	1.1	0.45	3.1	1.0	0.31	0.24	0.18
3.11.2010	80	15.6	5.8	0.85	6.5	1.1	0.42	0.11	0.24
12.11.2010	47	2.2	0.47	0.25	1.3	1.0	0.36	0.18	0.12
16.11.2010	46	3.6	0.74	0.33	1.7	0.91	0.29	0.20	0.36
17.11.2010	50	10.3	3.8	0.38	3.6	0.56	0.53	0.10	0.11

From the results it is obvious that the limit of 50 µg/m³ was exceeded 39-times from January to November 2010. In *Fig. 1* we can also see that the concentration of PM₁₀ was higher than 100 µg/m³ 4 times (2010-01-18, 2010-01-28, 2010-02-17 and 2010-02-18). The highest measured PM₁₀ concentration was 127 µg/m³ on 2010-02-18. The concentration of chloride and sodium is not directly related to the concentration of PM₁₀. It is also interesting that the ratio of chloride and sodium is usually 2:1 and 1:1. If the ratio is 2:1 the additional source for Cl⁻ is expected. In the addition, higher concentration of NO₃⁻ shows also on high pollution due to the anthropogenic source (most probably traffic). The highest concentration for NO₃⁻ was 20.0 µg/m³ on 2010-01-12. The lowest concentration of chloride that was measured was 0.11 µg/m³ (2010-10-11) and the highest was 6.89 µg/m³ (2010-01-28). The lowest determined concentration of sodium was 0.05 µg/m³ (2010-10-08) and the highest 3.99 µg/m³ (2010-01-27).

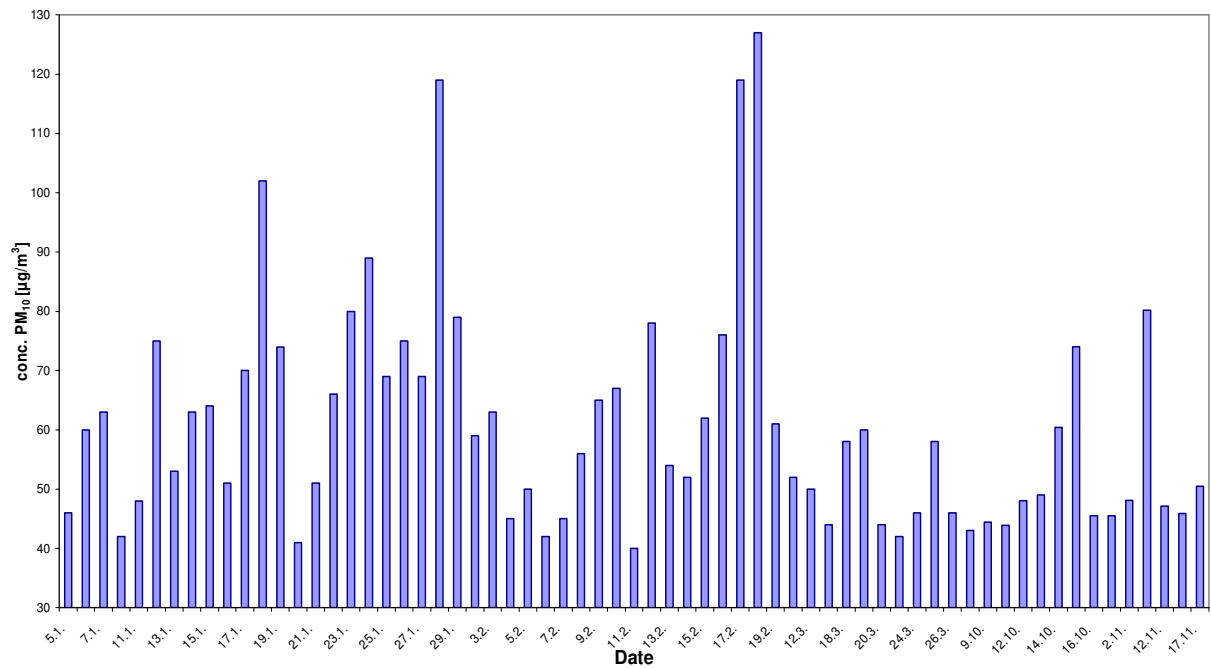


Figure 3: Concentrations of PM₁₀ (only PM₁₀ > 40 µg/m³) during sampling period from January to November 2010, sampling site Maribor.

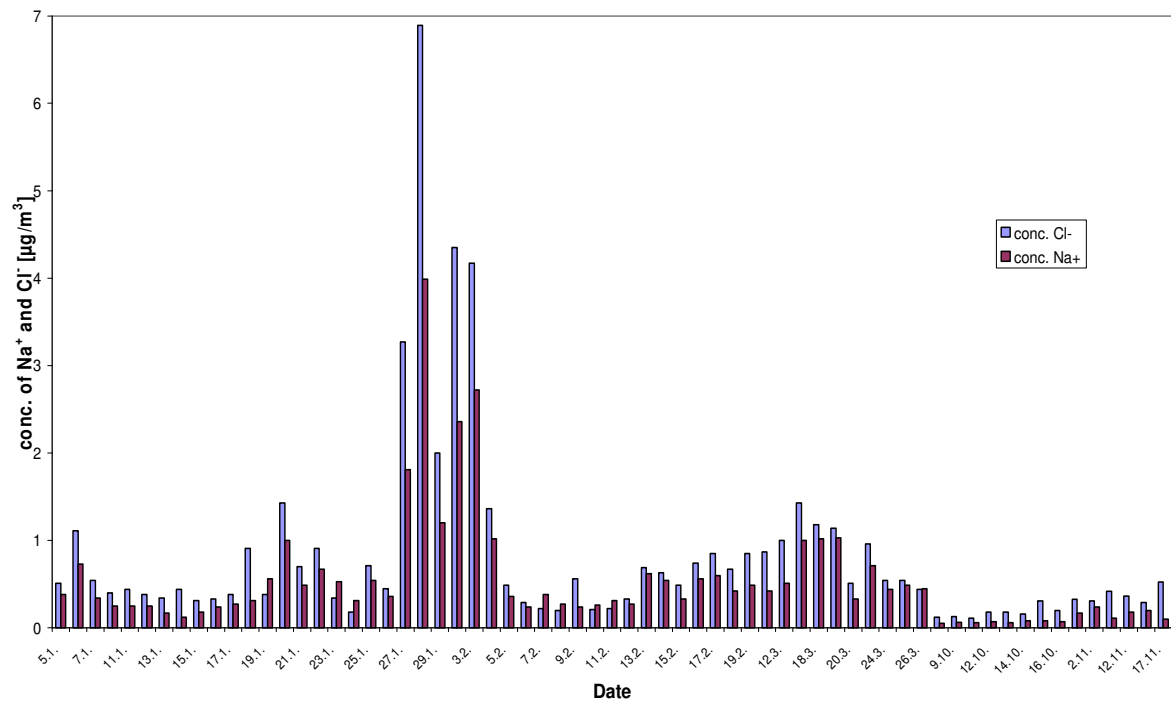


Figure 4: Concentrations of Na⁺ and Cl⁻ in the PM₁₀ (> 40 µg/m³) during the sampling period from January to November 2010, sampling site Maribor.

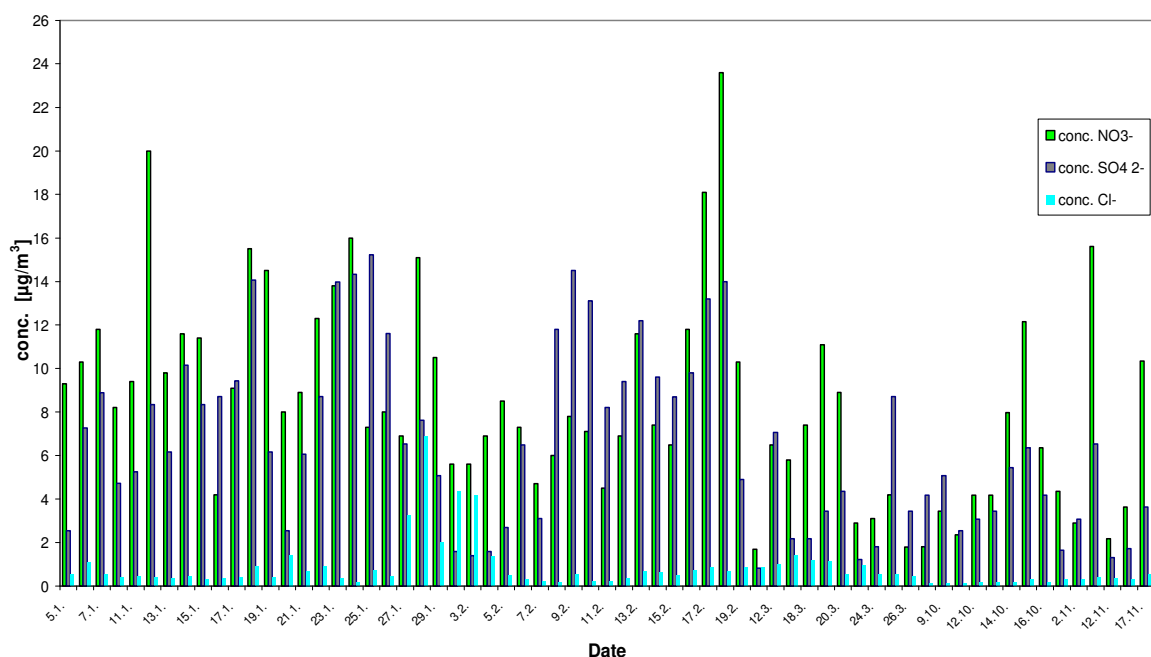


Figure 5: Concentrations of NO_3^- , SO_4^{2-} and Cl^- in the PM_{10} ($> 40 \mu\text{g}/\text{m}^3$) during the sampling period from January to November, 2010 sampling site Maribor.

The prevailing anions were NO_3^- and SO_4^{2-} . Among the cations, NH_4^+ was found at the highest concentration. Concentrations of Ca^{2+} , Mg^{2+} and Na^+ were similar.

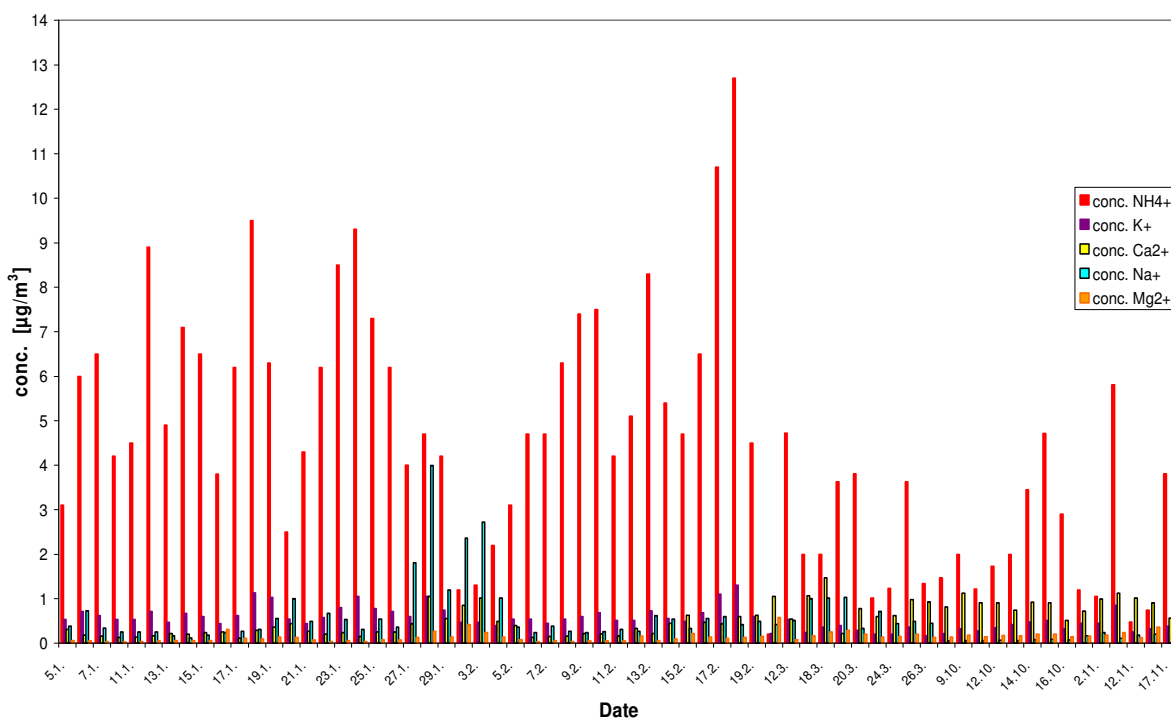


Figure 6: Concentrations of NH_4^+ , K^+ , Ca^{2+} , Na^+ and Mg^{2+} in the PM_{10} ($> 40 \mu\text{g}/\text{m}^3$) during the sampling period from January to November 2010, sampling site Maribor.

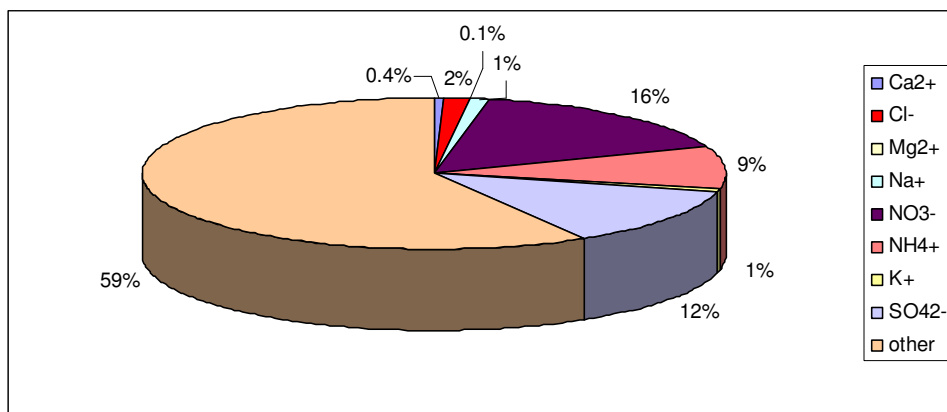


Figure 7: Portions (in %) of measured components (monthly average concentration) in PM₁₀ particles, January 2010

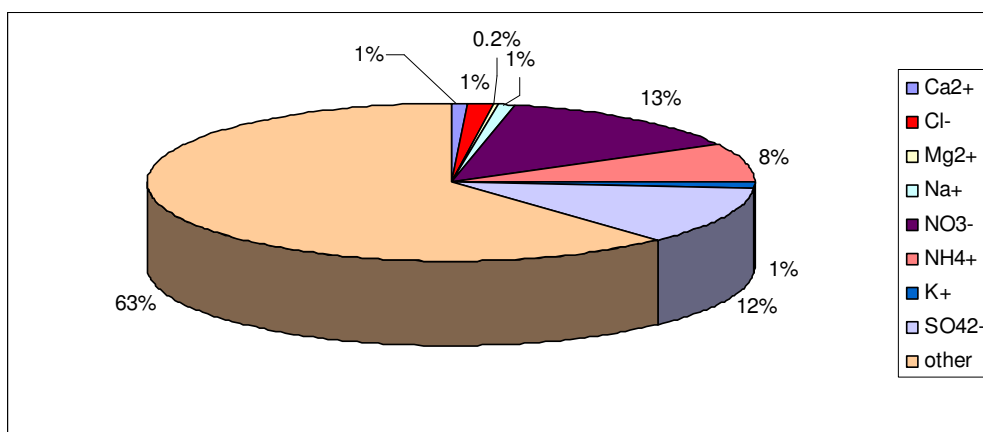


Figure 8: Portions (in %) of measured components (monthly average concentration) in PM₁₀ particle, February 2010

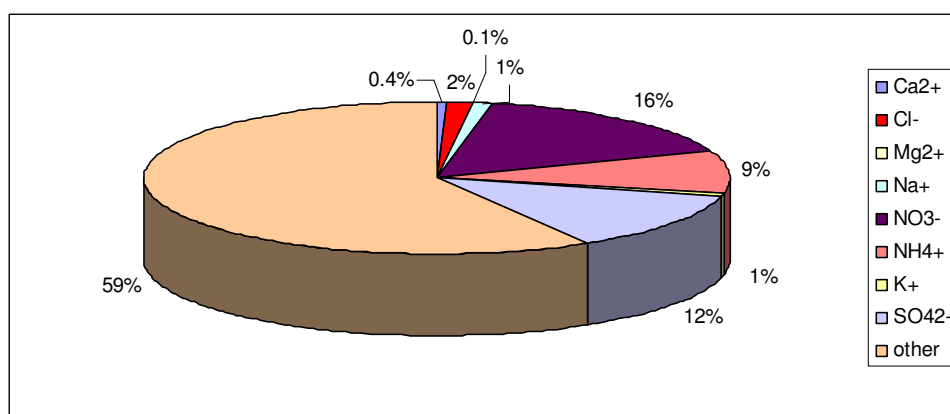


Figure 9: Portions (in %) of measured components (monthly average concentration) in PM₁₀ particles, March 2010

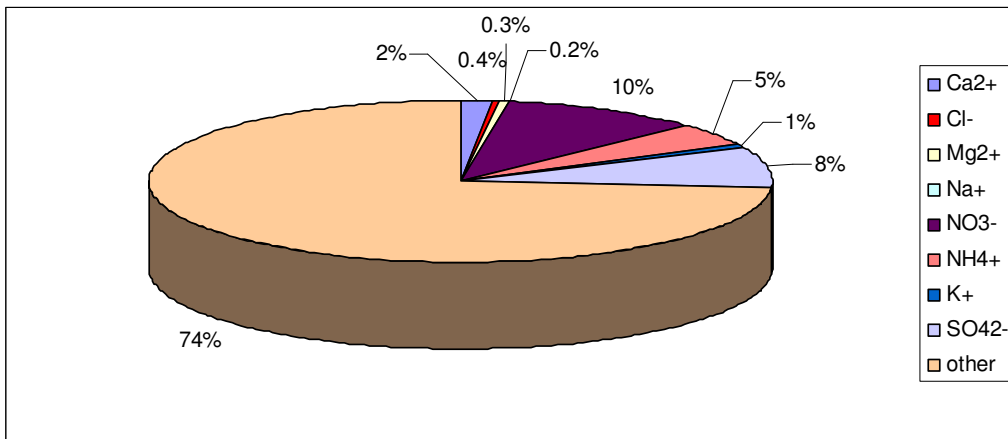


Figure 9: Portions (in %) of measured components (monthly average concentration) in PM_{10} particles, October 2010

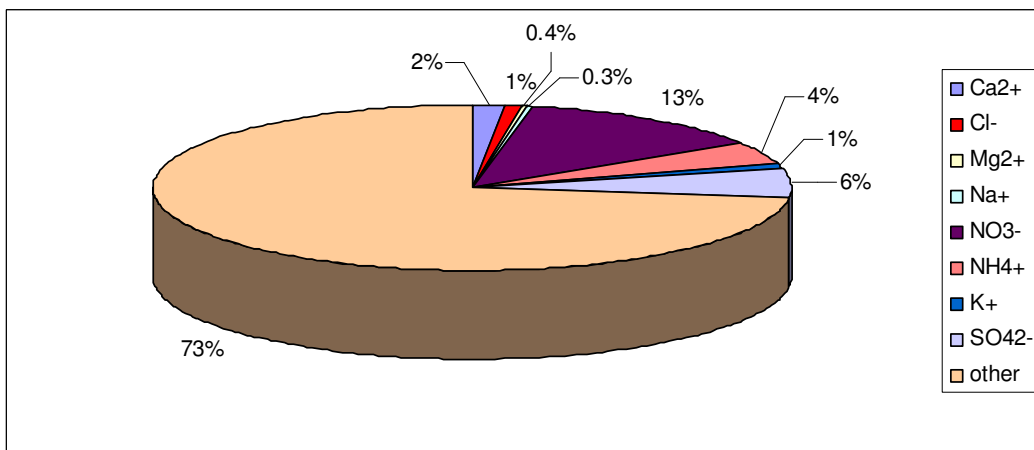


Figure 10: Portions (in %) of measured components (monthly average concentration) in PM_{10} particles, November 2010

It is obvious that for all five months in 2010 the major component in PM_{10} concentrations NO_3^- , SO_4^{2-} and NH_4^+ . It can also be concluded that in comparison with autumn during winter months higher concentrations of Cl^- (38 %) and Na^+ (21 %) were determined.

Table 6: Average concentrations of PM₁₀ and Cl⁻ concentrations for days with and without salting in Maribor: January, February, March, October and November 2010

Month		C _{PM10} (µg/m ³)	Chloride (µg/m ³)	
January	Salting	All days (9 days)	62	0.98
		PM ₁₀ >50µg/m ³ (6 days)	66	0.97
	No salting	All days	70	1.30
		PM ₁₀ >50 µg/m ³	75	1.42
February	Salting	All days (2 days)	59	0.48
		PM ₁₀ >50µg/m ³ (1 day)	78	0.42
	No salting	All days	68	1.26
		PM ₁₀ >50 µg/m ³	71	1.32
March	Salting	All days (1 day)	50	1.00
		PM ₁₀ >50µg/m ³	/	/
	No salting	All days	50	0.84
		PM ₁₀ >50 µg/m ³	59	0.96
October	Salting	All days	/	/
		PM ₁₀ >50µg/m ³	/	/
	No salting	All days	51	0.19
		PM ₁₀ >50 µg/m ³	67	0.23
November	Salting	All days*	50	0.53
		PM ₁₀ >50µg/m ³	/	/
	No salting	All days	55	0.34
		PM ₁₀ >50 µg/m ³	80	0.42
Total	Salting	All days	55	0.75
		PM ₁₀ >50µg/m ³	72	0.70
	No salting	All days	59	0.79
		PM ₁₀ >50 µg/m ³	70	0.87
Total (January. February. March)	No salting	All days	63	1.10
		PM ₁₀ >50 µg/m ³	68	1.20
Total (October. November)	No salting	All days	53	0.30
		PM ₁₀ >50 µg/m ³	74	0.30

* Only 1 day

The Cl⁻ concentrations for the selected PM₁₀ samples (> 40 µg/m³) for the autumn/winter period varied from 0.19 to 1.32 µg/m³ (Table 6). If all day or days with salting are taken into account, no big difference in Cl⁻ concentrations can be seen. But difference can be seen between the days with and without salting, especially on January (30 % higher on days without salting) and February (about 3-times higher on days without salting). Relation between the concentrations of Cl⁻ on days with no salting and days with salting has changed. In January, the ratio was 1:1.4, in February 1.2:3.0 and in March. 1:1. The impact of salting can be seen up to three days after salting of roads. It is clearly seen the difference between the months with salting (January, February, March 2010) and without salting (October, November 2010). Comparison between winter and autumn period shows about 4-times higher values for Cl⁻ for winter months.

Table 7: Average concentrations of PM₁₀ and Cl⁻ concentrations for all period (January, February, March, October and November 2010) days with and without salting in Maribor

Total	Salting	All days	55	0.75
		PM ₁₀ >50µg/m ³	72	0.70
	No salting	All days	59	0.79
		PM ₁₀ >50 µg/m ³	70	0.87

For the whole period the average chloride concentration is 1.4 % of the PM₁₀ on both days with salting and days without salting. But for October and November 2010 the concentration of Cl⁻ is about 0.6 % of the total PM₁₀.

For the Austria the average chloride concentration was 4 % of the total PM₁₀ on days with winter-salting and 3 % on days without salting.

5.1 Meteorology

5.1.1. Wind

At the measuring location in Maribor the wind direction was northwest with a very low speed (from 0 to 4 m/s²). As an illustration the wind rose for each month are present (Figure 11 – 15).

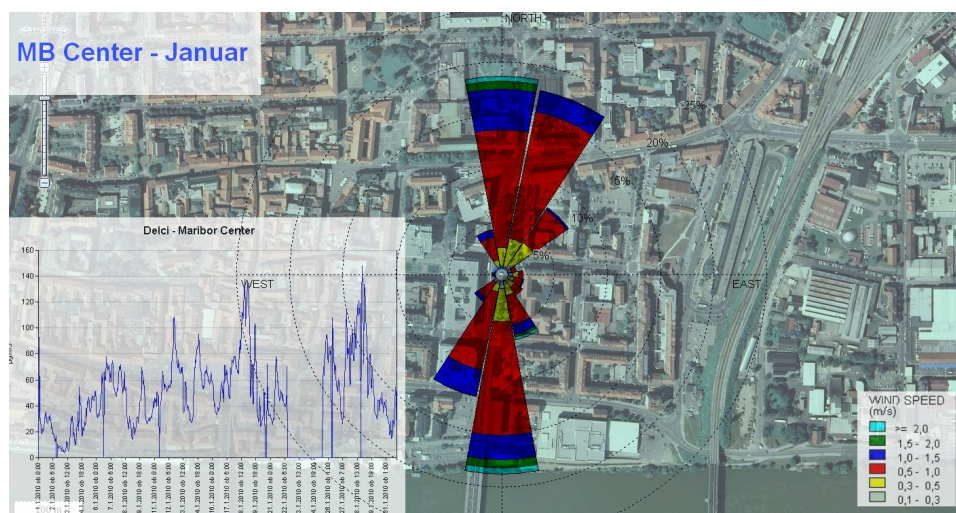


Figure 11: Wind rose for measuring site in Maribor in January 2010

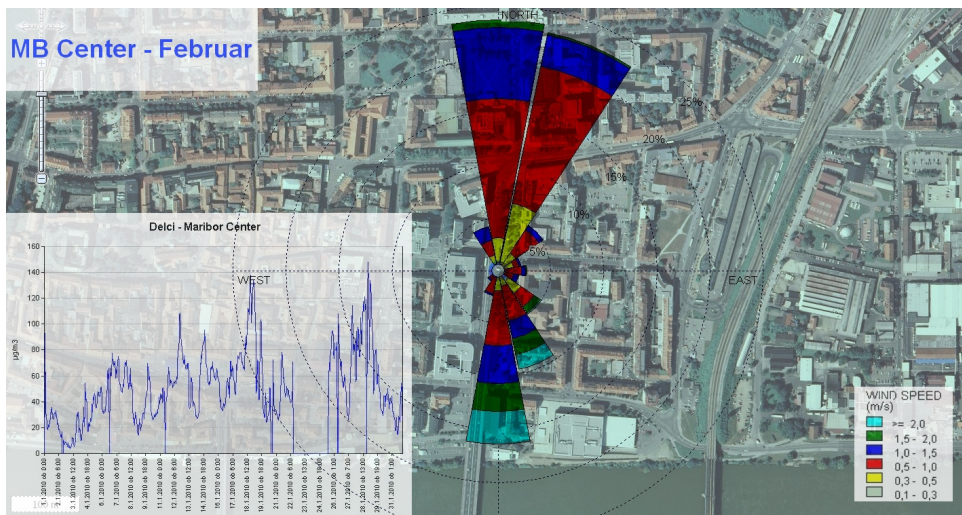


Figure 12: Wind rose for measuring site in Maribor in February 2010

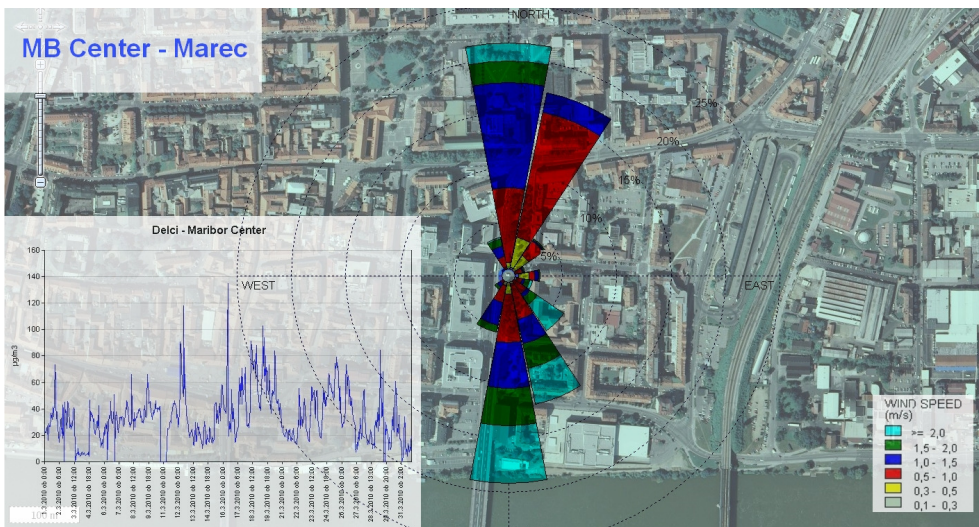


Figure 13: Wind rose for measuring site in Maribor in March 2010

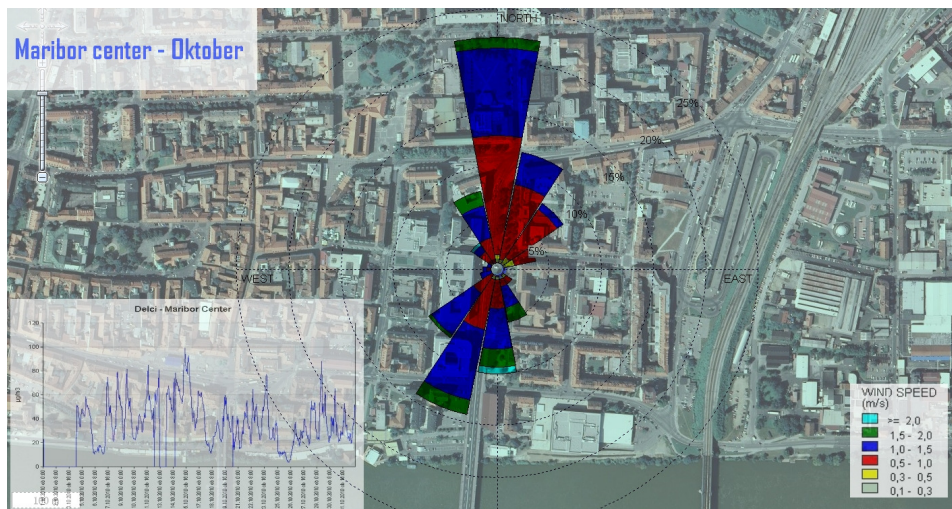


Figure 14: Wind rose for measuring site in Maribor in October 2010

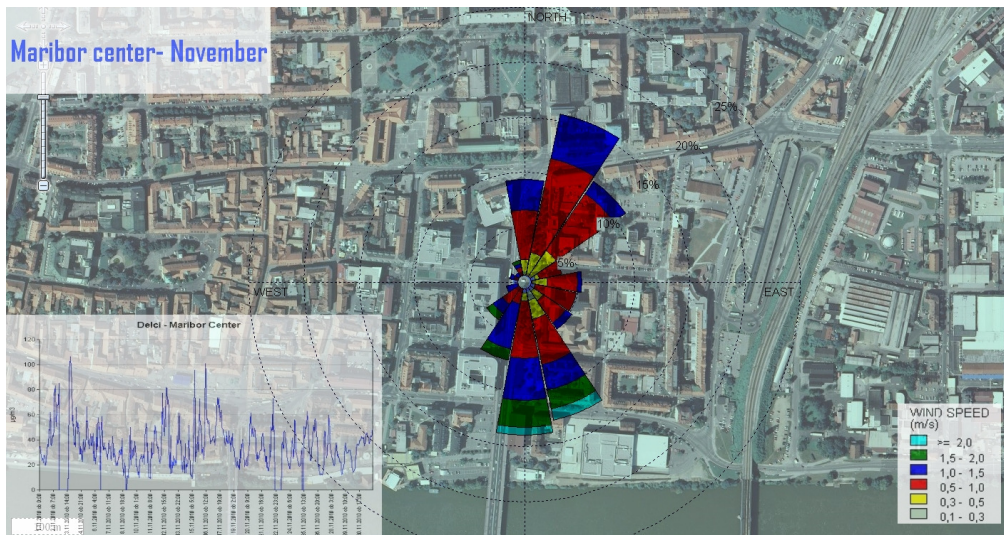


Figure 15: Wind rose for measuring site in Maribor in November 2010

Insets in Figures 11 – 15 present the 30 min concentrations of PM_{10} for each day of the month.³⁰

From this data (low wind speed) we can conclude that the contribution to Cl^- concentration from other source (also salting from other streets) is very unlikely.

5.1.2. Some important meteorological data (rain, snow, dry, humidity and temperature)

Table 7: Winter-salting, weather conditions, relative humidity and temperature at the measurement site in Maribor (for January, February, March, October and November 2010).

Date	c PM ₁₀	winter-salting	rain	snow	dry	RH		temperature	
	µg/m ³	number of services				over 80%	under 80%	over 0°C	under 0°C
4.1.2010	/	√√√√							
5.1.2010	46	√√							
6.1.2010	60	√√√√		√		√		√	
7.1.2010	63	√		√		√		√	
8.1.2010	42	√√√√√√√√√√							
9.1.2010	/	√√							
11.1.2010	48								
12.1.2010	75			√		√		√	
13.1.2010	53				√	√		√	
14.1.2010	63			√		√		√	
15.1.2010	64			√		√		√	
16.1.2010	51				√	√		√	
17.1.2010	70	√√		√		√		√	
18.1.2010	102				√	√		√	
19.1.2010	74				√	√			√
20.1.2010	41								
21.1.2010	51	√√√√√√							
22.1.2010	66	√√√√		√		√		√	
23.1.2010	80								
24.1.2010	89	√√√√		√		√		√	
25.1.2010	69								
26.1.2010	75	√√							
27.1.2010	69								
28.1.2010	119				√		√	√	
29.1.2010	79				√		√	√	
30.1.2010	/	√√							
2.2.2010	59				√		√	√	
3.2.2010	63								
4.2.2010	45								
5.2.2010	50								
6.2.2010	42	√√√√√√√√√√							
7.2.2010	45	√√							
8.2.2010	56								
9.2.2010	65	√							
10.2.2010	67	√√√√							
11.2.2010	40	√√√√√√√√√√							
12.2.2010	78	√√√			√	√		√	
13.2.2010	54			√			√	√	
14.2.2010	52				√		√	√	
15.2.2010	62				√	√		√	
Date	c PM ₁₀	winter-salting	rain	snow	dry	RH		temperature	

	$\mu\text{g}/\text{m}^3$	number of services				over 80%	under 80%	over 0°C	under 0°C
16.2.2010	76				√	√		√	
17.2.2010	119				√	√			√
18.2.2010	127		√			√			√
19.2.2010	61		√			√			√
23.2.2010	52				√		√		√
5.3.2010	/								
7.3.2010	/	√√							
10.3.2010	/	√√√√√√							
11.3.2010	/	√√√√√√							
12.3.2010	50	√√√√			√		√	√	
16.3.2101									
17.3.2010	44								
18.3.2010	58				√		√		√
19.3.2010	60				√		√		√
20.3.2010	44								
22.3.2010									
23.3.2010	42								
24.3.2010	46								
25.3.2010	58				√		√		√
26.3.2010	46								
6.10.2010	/		√						
7.10.2010	/		√						
8.10.2010	43				√	√			√
9.10.2010	44				√	√			√
11.10.2010	44				√		√		√
12.10.2010	48				√		√		√
13.10.2010	49				√	√			√
14.10.2010	60				√		√		√
15.10.2010	74				√	√			√
16.10.2010	46				√		√		√
17.10.2010	/		√						
18.10.2010	/		√						
20.10.2010	/		√						
21.10.2010	/		√						
23.10.2010	46				√		√		√
2.11.2010	48				√	√			√
3.11.2010	80				√	√			√
4.11.2010	/		√						
8.11.2010	/		√						
9.11.2010	/		√						
11.11.2010	/		√						
12.11.2010	47				√		√		√
16.11.2010	46				√	√			√
17.11.2010	50	√			√	√			√

6. CONCLUSIONS

- Contribution of winter salting is certainly evident:

- However not immediately but often after salting (2 – 3 days).
- The main meteorological conditions which from the Cl^- concentration are: dry whether. low relative humidity and wind direction.
- Effect of salting is evident from:
 - Usually higher concentration of PM_{10} .
 - The higher concentration of Na^+ and Cl^- .
 - The ratio between Na^+ and Cl^- was between 1:1 and 1:3.3
 - The highest measured concentration of Cl^- was $6.9 \mu\text{g}/\text{m}^3$.
- So, it can be conclude that the situation in Maribor was different to that in Austria. The reason could be different way of salting, they used brine or solution. while in Slovenia NaCl in solid form was used. As a result, in Austria tee effect of salting can be noticed in higher PM_{10} and Cl^- immediately after salting, while in our case after 1-2 days, mostly under dry conditions.

For the statistical evaluation for source apportionment we have insufficient data (at least 100 data are necessary). It is planned to continue the project because we expect more results for December 2010. I hope that the whole project will help to demonstrate that the effect of salted roads is actual and only source of the contribution to the PM_{10} concentration.

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