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Road salt emissions: A comparison of measurements and modelling using the NORTRIP road dust emission model

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HIGHLIGHTS

- For the first time emissions of road salt have been modelled.
- The model successfully reproduces many of the observational datasets.
- Salt emissions in PM10, during winter, are as high as exhaust emissions.
- A relationship between road salt application and concentrations is presented.

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ABSTRACT

De-icing of road surfaces is necessary in many countries during winter to improve vehicle traction. Large amounts of salt, most often sodium chloride, are applied every year. Most of this salt is removed through drainage or traffic spray processes but a certain amount may be suspended, after drying of the road surface, into the air and will contribute to the concentration of particulate matter. Though some measurements of salt concentrations are available near roads, the link between road maintenance salting activities and observed concentrations of salt in ambient air is yet to be quantified. In this study the NORTRIP road dust emission model, which estimates the emissions of both dust and salt from the road surface, is applied at five sites in four Nordic countries for ten separate winter periods where daily mean ambient air measurements of salt concentrations are available. The model is capable of reproducing many of the salt emission episodes, both in time and intensity, but also fails on other occasions. The observed mean concentration of salt in PM_{10}, over all ten datasets, is 4.2 μg/m^3 and the modelled mean is 2.8 μg/m^3, giving a fractional bias of −0.38. The RMSE of the mean concentrations, over all 10 datasets, is 2.9 μg/m^3 with an average R^2 of 0.28. The mean concentration of salt is similar to the mean exhaust contribution during the winter periods of 2.6 μg/m^3. The contribution of salt to the kerbside winter mean PM_{10} concentration is estimated to increase by 4.1 ± 3.4 μg/m^3 for every kg/m^2 of salt applied on the road surface during the winter season. Additional sensitivity studies showed that the accurate logging of salt applications is a prerequisite for predicting salt emissions, as well as good quality data on precipitation. It also highlights the need for more simultaneous measurements of salt loading together with ambient air concentrations to help improve model parameterisations of salt and moisture removal processes.

1. Introduction

Non-exhaust traffic induced particle emissions are known to

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contribute significantly to the total concentrations of inhalable airborne particulate matter in the size range <10 μm (PM_{10}), e.g. Amato et al. (2014), Mathissen et al. (2012), Bukowiecki et al. (2010), Gustafsson et al. (2008), Norman and Johansson (2006) and Boulter (2005). This is particularly true in Nordic countries where studded tyres are used during winter. Non-exhaust PM_{10} emissions are composed of a number of wear sources (road, tyre and brake) and may also contain emissions from traction sanding, from deposited material from construction work, from road side soil sources or from deposition of atmospheric PM. Another source of traffic induced PM is road salt, applied as a de-icing agent during winter.

Various measurements combined with receptor modelling (Wåhlin et al., 2006; Hagen et al., 2005; Richard et al., 2011; Ducret-Stich et al., 2013; Gianini et al., 2012) have indicated that the contribution of road salt to PM_{10} concentrations can be significant if dry residual salt is left on the road after de-icing is carried out. Contributions of salt vary but during winter months this can range from 10% to 35% of the average PM_{10} concentration. In some cases (Wåhlin et al., 2006; Hagen et al., 2005) daily mean salt concentrations can contribute to exceedances of the EU daily mean limit value for PM_{10} (>50 μg/m^3). In recognition of the possible contribution of road salt, and the importance of salting for traffic safety, the EU air quality directive (EC, 2008) allows the contribution of salt, and traction sand, to be discounted if it contributes to exceedances of the limit values. Guidelines for determining the road salt contribution, that are based on daily mean measurements of chloride, are provided in EC (2011).

In general road salting is considered necessary for road safety and accessibility. Large amounts of salt are used every winter season in Nordic countries. In the four Nordic countries of Sweden, Norway, Finland and Denmark more than 500,000 tonnes of salt is used each year (NVF, 2014). Salt is also seen as an environmental pollutant, having a negative effect on vegetation, surface and ground water aquifers as well as eco-systems in proximity to salted roads. Efforts are therefore made to reduce the use of salt by more efficient application. Most efforts to date to study salt processes involves the drainage or spray of salt from the road surface (e.g. Blomqvist and Johansson, 1999; Blomqvist et al., 2011). Modelling of the suspension of dry salt, often referred to as salt 'blow off', has to date not been attempted.

Determining the amount of salt in ambient air concentrations is generally carried out by the collection of filter samples and elemental or ionic analysis of chlorine (Cl) and/or sodium (Na) (Wåhlin et al., 2006; Laupsa et al., 2008; Richard et al., 2011; Ducret-Stich et al., 2013; Gianini et al., 2012). These types of measurements are limited to one or perhaps two sites within a city, if any at all. It is therefore of interest to assess the possibility of modelling road salt emissions and the contribution of this source to PM_{10} concentrations. To date no road salt emission modelling has yet been carried out.

The NORTRIP road dust emission model (Denby et al., 2013a, 2013b) is capable of calculating road salt emissions. It is a coupled road dust and surface moisture model that describes a range of processes related to the generation and removal of dust and salt on the road surface as well as their subsequent suspension and emission. Suspension and retention of dust and salt is governed by the road surface moisture and this is described using a mass and energy balance approach where the surface moisture accumulates through precipitation, condensation and wetting (by road maintenance activities). Moisture is removed through evaporation, drainage and vehicle spray processes.

Salt is included in the model as a mass source and can be in solution or in solid form (oversaturated). Salt removal processes are dominated by drainage but wet removal through vehicle spray or snow/slush removal can also be significant. Both normal de-icing salt (NaCl) as well as the dust binding solutions of magnesium chloride ($MgCl_2$), calcium chloride ($CaCl_2$) and calcium magnesium acetate (CMA) are included in the model. In Denby et al. (2013b) the model was compared to PM_{10} measurements at a number of sites in Nordic countries and salt emissions were briefly reported. Apart from this no modelling studies of salt emissions from roads has, to date, been carried out.

In this paper we apply the NORTRIP model to ten datasets at five locations where ambient concentrations of salt have been measured near roads and where some information is available concerning the winter road maintenance salting activities. These datasets are from the four Nordic cities of Oslo (Norway), Stockholm (Sweden), Helsinki metropolitan area (Finland) and Copenhagen (Denmark) and as such cover a range of conditions. Some of these sites have already been used for validation of the NORTRIP model in relation to road dust emissions (Denby et al., 2013a, 2013b) but no details concerning the salt contribution were given.

We first describe the measurements and sites and then provide a summary of the processes described by the NORTRIP model that relate to salt loading and emissions. Results of the model calculations and a comparison with observations is then made. An analysis of the surface salt mass balance is carried out and a number of sensitivity tests are also provided to assess model parameter and input data uncertainties. We close with discussion and conclusions.

2. Measurement sites and data

Five kerbside measurement sites in four different countries are used in this study. At all these sites, filter samples of PM_{10} were collected and analysed for Sodium (Na) and in some cases also Chlorine (Cl) to provide daily mean concentrations. All sites also measured hourly or daily mean concentrations of NO_X and PM_{10}. The method of chemical analysis varied from site to site and is specified in the following sections. Only the Copenhagen site, which is the most exposed to sea salt contributions, had an additional salt measurement at an urban background site that could be used to subtract urban background concentrations. Regional background salt concentrations were used to remove sea salt contributions for the Hämeentie site in Helsinki.

The three sites Stockholm, Copenhagen and Oslo have already been used to verify the road dust emission model NORTRIP (Denby et al., 2013b) though the periods covered differ. In Table 1 information concerning the five sites is provided. We briefly describe each of the sites and any special characteristics of the measurement periods.

2.1. Hornsgatan (Stockholm)

Hornsgatan is a four lane street canyon site in central Stockholm with approximately 20,000 vehicles per day and a relatively low percentage (3%) of heavy duty vehicles. Average speeds are around 44 km/h. PM sampling was made at 3 m above the road surface at the Northern side of the street using a sampler with a PM_{10} (Leckel SEQ 45/50) inlet compliant with the EN12341 standard of the European Committee for Standardization (CEN). The filters (Pallflex, EMFAB TX 47 mm, borosilicate glass bonded with PTFE) were analysed for inorganic ions using ion chromatography.

At this site CMA (Calcium Magnesium Accetate) is applied for dust binding and regular cleaning is also carried out. Deicing salt is applied around 50 times per season with an estimated 10 g/m^2 for each application. A limited number of road surface samples were also taken during the course of the winter seasons using the wet dust sampler (Jonsson et al., 2008). These were aimed primarily at assessing the dust loading of the road surface but they were also

Table 1
Summary site description and data availability at the five sites used in this study. When multiple years are used for a particular site then individual information is shown per year if information significantly differs between the years, otherwise an average is shown.

Site name	Hornsgatan	HC Anderson Boulevard (HCAB)	Riksvei 4 (RV4)	Kehä III	Hämeentie
City, country	Stockholm, Sweden	Copenhagen, Denmark	Oslo, Norway	Helsinki, Finland	Helsinki, Finland
Periods assessed	2 winter periods of 5 months each 2012–2013	4 winter periods of 4 months 2010–2013	2 winter periods of 4 and 6 months 2004–2005	1 winter period of 3 months 2013	1 winter period of 4 months 2014
Number of days assessed	151	121	2004: 121 2005: 181	90	120
Traffic ADT (veh/day)	20 000	57 000	42 000	32 000	14 500
Heavy duty vehicle fraction (%)	3	3	5	10	25
Average speed (km/hr)	44	21	69	83	36
Number of lanes	4	6–7	4	4	4
Number of salting operations	2012: 56 2013: 47	2010: 30 2011: 41 2012: 21 2013: 52	2004: 113 2005: 119	41	17
Number of dust binding operations	2012: 31 2013: 38	0	0	0	10
Total salt mass applied to street during the study period (tonne/km)	2012: 11 2013: 8	2010: 11 2011: 14 2012: 8 2013: 19	2004: 39 2005: 37	5.7	2.6
Number of snow ploughing operations (modelled)	2012: 0 2013: 9	2010: 2 2011: 6 2012: 1 2013: 0	2004: 9 2005: 3	35	4
Quality of road maintenance salting data	Timing known and salt mass estimated at 10 g/m^2 per application	Timing known within a 6 h window of time, salt mass estimated at 17 g/m^2 per application	Complete log data with specified salt mass but from another road so can deviate	Timing known and salt mass estimated at 10 g/m^2 per application	Timing known within a day (assumed to occur at 5 a.m.) and salt mass estimated at 10 g/m^2 per application
Number of daily mean measurements of ambient air salt concentrations	2012: 29 2013: 51	2010: 111 2011: 103 2012: 103 2013: 70	2004: 40 2005: 40	21	83
Background measurements of ambient air salt concentrations	No	Yes, 3 km from traffic site	No	No	Yes, regional background estimate available
Filter sample analysis	Ionic analysis	Only Na	Elemental and ionic analysis	Ionic and elemental analysis	Ionic and elemental analysis
Measurements of salt mass on the road	2012: 6 2013: 7	None	None	None	None
Meteorological data source	Temperature and relative humidity in street canyon. Wind, precipitation and radiation at roof top.	Temperature, humidity, radiation and wind from station 3 km away. Precipitation from stations 1.5/2 km away	Temperature, humidity and wind from station 2 km away. Radiation from station 30 km away.	Temperature, humidity, precipitation and wind from station 15 km away. Radiation from station 11 km away.	Temperature, humidity, precipitation, cloud cover and wind from station 1 km away. Radiation from 3 km away.
Precipitation (mm)	2012: 174 2013: 152	2010: 141 2011: 128 2012: 191 2013: 93	2004: 178 2005: 128	173	174

analysed for ions and for acetate, to identify salt and the dust binder CMA. For this site measurements of the salt loading are also presented. NO_x and PM-exhaust emission factors for Hornsgatan are based on the observed fleet composition, driving speed and driving conditions and have been estimated using the HBEFA 3.1 emission model (HBEFA, 2015).

2.2. HCAB (Copenhagen)

HCAB is a monitoring station at H.C. Andersens Boulevard, a busy 6–7 lane (one left turn lane close to monitoring location) road in the centre of Copenhagen. HCAB is, with 57,000 veh/day, one of the busiest roads in Copenhagen and the station with the highest NO_2 and PM_{10} concentrations in the National Monitoring Programme in Denmark (Ellermann et al. 2015). The street has 4–5 floor buildings near the monitor station and is open towards a park area (Tivoli Gardens) at the opposite side. The signed speed is 50 km/h but due to congestion and traffic lights the average travel speed (measured by GPS recordings) is only about 21 km/h. Recordings of the salting activities were obtained from Copenhagen road administration. These provided an approximately 6 h window in which activities occurred. Salt mass was estimated to be 17 g/m^2

per application. Background measurements for air pollution, temperature, radiation, wind and humidity are taken from a roof station (H.C. Ørsteds Institute) 3 km from HCAB. Precipitation data were obtained from two different stations operated by Danish Metrological Institute 1.5 km and 2 km from the site. Manual traffic counts provide hourly variation of traffic and the distribution into vehicle categories. This is further distributed into engine size, age, Euro class and fuel type based on the Danish National vehicle register. NO_X and PM-exhaust emissions are estimated using COPERT IV (COPERT, 2014) emission factors, including 'real world' adjustment for diesel cars in EURO 5 + 6.

2.3. RV4 (Oslo)

Filter sample measurements were taken at RV4 as part of an experiment into the impact of speed signage on road dust emissions (Hagen et al., 2005). Speed signage changed in autumn 2004 from 80 to 60 km/h on this road. The station is close to a major arterial road in Oslo near the Akershus hospital. A background station for PM concentrations, no chemical analysis was carried out, was placed in the hospital grounds. At this time experiments were also being carried out by the road authorities in other parts of Oslo to assess the use of $MgCl_2$ as a de-icer and dust binder. No $MgCl_2$ was used on RV4 during the measurement period but information concerning winter road salting on another stretch of road in Oslo was collected during the road authority experiments and these data are used here. PM samples were taken daily and analysed for a range of ions and metals for receptor modelling applications (Laupsa et al., 2008). A selection of the available samples was made so the analysis is not continuous. For RV4 the NO_x and PM-exhaust emission factors are based on a report from Hagman et al. (2011).

2.4. Kehä III (Helsinki metropolitan area)

The air quality monitoring station is located on the shoulder of Kehä III, one of the three major ring roads in the Helsinki metropolitan area, in the city of Vantaa. The speed limit (80 km/h) and average driving speed (83 km/h) is the highest of all the study sites. The proportion of heavy duty vehicles was also quite high (10%). Daily PM_{10} filter samples were collected between 26 February and 17 May in 2013. The salt analysis was only performed on 21 selected samples since the main objective of the measurements was to evaluate PM_{10} sources during the days when daily mean PM_{10} concentrations exceeded 50 $\mu g/m^3$. After extraction of the filters with water, Na was analysed with ion chromatography (IC) and Cl with inductively coupled plasma mass spectrometry (ICP-MS). Dust binding was not used at this measurement site.

2.5. Hämeentie (Helsinki metropolitan area)

The air quality monitoring station at Hämeentie is located at a busy street canyon in Helsinki centre. Hämeentie is characterized with unusually high proportion of heavy duty vehicles (25% of the traffic flow) since it is a major route for city buses. Daily PM_{10} filter samples were collected between 28 February and 31 May in 2014. The Na and Cl analysis of the samples was performed in a similar way as described for Kehä III. For this site regional background salt concentrations were available and have been subtracted from the dataset to remove sea salt contributions. NO_X and PM-exhaust emission factors for both Kehä III and Hämeentie are based on the observed and modelled fleet composition, driving speed and driving conditions and have been estimated using the HBEFA 3.1 emission model (HBEFA, 2015).

3. Modelling

The NORTRIP model is a coupled road dust/salt and moisture model. It is based on the physical processes that govern the mass balance of water/ice/snow and of dust/salt. Included in the model are elements governing the energy balance of the road surface, the surface temperature, the impact of salt on vapour pressure and melting and the suspension and emissions of dust/salt. The model contains all the elements of a typical 'road weather model' in order to calculate the road surface conditions that determine if emission or retention of particles occurs. Input data required by the model includes hourly traffic and meteorological data as well as road maintenance activity data for salting, dust binding, cleaning and snow ploughing.

The model has been previously described in Denby et al. (2013a, 2013b) where the focus was on road dust emission modelling, though salt was already included at that stage. In this current study we focus on the salt and its emissions. We do not look closely at the impact of dust binding here as this will be the subject of a further study. In order to correctly model salt emissions the following three main intermediate steps must be attained:

1. The salt loading on the road surface must be correctly predicted, requiring accurate salt application data as well as process descriptions of salt removal by drainage, vehicle spray and snow removal processes.
2. The surface moisture and retention conditions must be correctly predicted, requiring accurate input data for precipitation as well as process descriptions for drainage, vehicle spray, snow removal, salt impact on phase changes and surface temperature.
3. The suspension of salt by traffic in the PM_{10} size fraction must be correctly described, requiring accurate input data for traffic as well as process descriptions for traffic suspension and size distribution of salt.

Attaining the above is demanding for a model and there can be a significant amount of uncertainty in each of these steps and process descriptions. The sensitivity of the model to a number of these processes and input data will be assessed in Section 5.3. In this section we briefly summarise the important aspects of the model in regard to the salt emissions. In general we refer the reader to Denby et al. (2013a, 2013b) for a full description.

3.1. Salt application

The application of salt to the road surface is specified as input data to the model and is usually derived from road maintenance logs, though the amount of salt for each application is rarely provided and estimates are required. The mass of salt (g/m^2) and the amount of water (kg/m^2 or mm) is given as input to the model for any specific hour, Section 3.4 Denby et al. (2013a). Salt is assumed to be applied over the entire available road surface and with no loss of salt due to 'bounce off'. The quality of the data used in this study varies (see Table 1) so uncertainty in these input data will impact on the results. Sensitivity tests are carried out for this parameter.

3.2. Drainage removal

Drainage removal is described in Section 3.6 Denby et al. (2013a). Rain or melt water production is assumed to occur at a constant rate during an hour that continually flushes the road surface reservoir of water, with a maximum water depth specified by the parameter $g_{drainable} = 0.5$ mm. The mixing efficiency of salt is assumed to be unity, i.e. completely mixed, when salt is in solution. The parameter $g_{drainable}$ is important for the salt removal as it controls the amount of water left on the road after drainage as well as the depth of the reservoir whilst draining. In reality drainage will depend on the down road slope, the cross-fall slope and the surface macro-texture, i.e. the roughness of the surface. However, this information is not available at the sites so the threshold depth is set to be constant for all roads. Sensitivity tests are carried out for the drainage parameter and the efficiency of salt mixing.

3.3. Spray removal

Vehicle spray removes water, snow and salt mass in solution from the road surface. It is vehicle speed and vehicle type dependent. Spray will be a major removal source of water and salt on higher speed roads. The method for describing spray has been slightly updated from that given in Section 2.5 of Denby et al. (2013b) as it now includes a threshold vehicle speed ($V_{thresh,spray} = 20$ km/h) below which no spray occurs, but otherwise the same quadratic speed dependence is retained. Apart from the speed dependence the essential user defined parameter is the spray rate (fraction of surface water or snow removed with the passage of each vehicle) which is given as $f_{0,spray} = 1 \times 10^{-4}$ (veh^{-1}) for light duty vehicles at the reference speed of 70 km/h. Heavy duty vehicles are given a spray rate 6 times larger and snow removal by spray is ten times smaller.

3.4. Removal through cleaning and snow ploughing events

Salt removal by cleaning and snow ploughing is simply represented in the model. A user specified fraction of the existing salt load on the road surface is removed during these events, Section 2.6 Denby et al. (2013b). No observational data is available to specify what this fraction may be but we specify the fraction of salt removed to be 0.2. When flushing is used for cleaning and the quantity of water applied is above the drainage threshold of 0.5 mm (Section 3.2) then both water and salt will be removed through the drainage process.

3.5. Suspension

Suspension is both a removal process for the road salt loading and the means by which salt is emitted to the air. Suspension of dry salt occurs when the road dries out below a particular level of surface moisture ($g_{retention\text{-}min} < 0.04$ mm). The total suspension will depend on the number of vehicles and the suspension rate per vehicle. The suspension rate per vehicle is, in turn, dependent on vehicle type (light or heavy) and vehicle speed, Section 3.5 Denby et al. (2013a).

The suspension rate for road dust was analysed in Denby et al. (2013a) and found to be significantly lower than that found in other studies (e.g. Patra et al., 2008). This is because the suspension rate described in most experiments is related to suspension from the wheel tracks, which are quickly emptied of dust or salt. In the model this type of suspension is described by a ‘direct emission’ for road dust, Section 3.8 Denby et al. (2013a). The longer time scales for suspension observed for road dust emissions in Section 4.1 Denby et al. (2013a) are more related to suspension through vehicle turbulence and the rate of vehicle meandering over the road. However, since the model is a single track model, i.e. it does not distinguish between in and between wheel tracks, then salt is assumed to also follow the longer time scale suspension rate. The model then, in its current form, cannot adequately predict the suspension of dry salt immediately after distribution, as in Patra et al. (2008). However for salt that has crystallised on the surface after being in solution this suspension description should be adequate. Sensitivity tests are carried out on this parameter.

3.6. Fraction of emitted salt in PM_{10}

The amount of suspended salt in the PM_{10} fraction is unknown. Currently the PM_{10} fraction of road wear dust is set at 28% in the model, based on previous modelling studies and laboratory and field measurements (Snilsberg at al., 2008). Since there are no size specific measurements of re-crystallised road salt it is not possible to know the size distribution of suspended dry salt. We assume that 28% of the suspended salt is in the size range <10 μm, as with dust.

3.7. Vapour pressure and dust binding

The dust binding potential of the salt and acetate solutions is implemented in the model by allowing them to change the surface vapour pressure, see Section 2.9 Denby et al. (2013b). A vapour pressure deficit and change in freezing point is specified for salt solutions under saturation based on data from the literature. The vapour pressure deficit essentially allows condensation of water vapour from the atmosphere onto the salt or acetate, keeping the surface moist even under relatively dry atmospheric conditions. Sensitivity tests for dust binding are carried out.

3.8. Calculating concentrations from emissions

In all results presented in this study use is made of observed NO_X concentrations and estimated NO_X emission factors to convert emissions to concentrations. This avoids the need for dispersion models and is considered to be more accurate, providing the NO_X emission factors are well defined. This approach is described in Section 3.9 Denby et al. (2013a).

4. Methodology

In all ten different winter periods for five different sites are modelled using NORTRIP. All these sites have a number of ambient air measurements of daily mean salt available for comparison. A reference run is made for all sites where all model parameters are the same. In addition a number of sensitivity tests are carried out related to model parameterisations and input data. This is limited to two sensitivity runs per dataset.

Time series plots (Section 5.1)

The results are presented as time series of daily mean PM_{10} salt concentrations and as salt loading on the surface for each site. Included in these time series is information on the salt application and the precipitation, in order to understand the modelled emissions and loadings. Both the reference and the two sensitivity calculations are shown for salt concentrations and loading. The salt application and precipitation shown are for the reference run only.

Statistical summaries (Section 5.2)

For each reference run and the two sensitivity runs per dataset the modelled mean and the correlation (R^2) is calculated for days when observations are available. For the sites HCAB and Hämeentie urban and regional background concentrations of salt are available, respectively, so that net observed salt concentrations can be determined. In all other cases the comparison is with the total observed salt which will include any urban or regional background.

Sensitivity analysis (Section 5.3)

For each site and dataset two additional calculations are carried out to assess the sensitivity of the model to a range of parameters and input data. The sensitivity tests can be grouped into four separate types.

1 Sensitivity to precipitation input data
2 Sensitivity to salt input data
3 Sensitivity to uncertain moisture related parameters and processes
4 Sensitivity to uncertain suspension related parameters

The choice of sensitivity for each dataset is intended to show some specific feature. For example the HCAB dataset comprised of a standard precipitation dataset from a site located near the former Royal Veterinary and Agricultural University. An alternative site (Botanic Gardens) was also available. When these two datasets differed significantly then sensitivity of the model to the alternative precipitation data was assessed (HCAB, 2011–2013). A short description of each of the sensitivity tests is given in Table 2.

Salt mass balance and contribution to emissions (Section 5.4)

For each dataset an analysis is made of the salt mass balance, for the reference calculation. The fraction of total applied salt that is emitted in PM_{10} is also assessed.

5. Results

5.1. Time series of model reference and sensitivity calculations

In this section plots of modelled and measured salt concentrations and salt loadings are given for each of the sites and each of the years. A short summary of each calculation is provided.

5.1.1. Hornsgatan (Stockholm)

Two years of data are shown for Hornsgatan (Figs. 1 and 2) which also includes occasional measured road surface salt loading. Specific to Hornsgatan is also the use of CMA as a dust binder. Levels of NaCl from 1 to 2 $\mu g/m^3$ can be seen in May, long after the last salting operation in both years and this is assumed to represent background levels, presumably due to sea salt contributions. For 2012 the observational data is limitted but compares well with the model calculations.

In 2013 the only significant observed salt contribution to PM_{10} is during March 2013 (Fig. 2). This is to some extent reproduced by the model but a significant peak (22 March 2013) is not reproduced until several days later. When this peak is observed then the model predicts wet surface conditions for several days after the snow fall on 18–20 March. A sensitivity test was carried out by removing this snow fall but the strong peak is not reproduced. For both years a sensitivity test was carried out by removing the dust binding in the model. This had a significant impact on the salt emissions in both years.

5.1.2. RV4 (Oslo)

The results for Oslo are shown in Figs. 3–4. In 2004 (Fig. 3) we see a very good correspondence between modelled and observed salt concentrations, with the model reproducing the timing well but with a slight underestimation of the concentrations. In 2005 (Fig. 4) many of the observed salt peaks are seen in the model but they tend to be underestimated when compared to the measurements. It should be re-iterated that the model salt concentrations are net concentrations, contributed from the local road, whilst the measured concentrations represent the total contribution from the Oslo urban region. For this particular site roughly 40% and 50% of the kerbside PM_{10}, for the years 2004 and 2005 respectively, can be attributed to the urban background. We would then expect, if salt is also emitted from other roads, that the modelled salt should under predict the measured concentrations.

The sensitivity tests for the 2004 data, doubling and halving of the water spray rate indicate sensitivity to this parameter. Salt loading and emissions increase when this removal mechanism is reduced and decrease when spray removal is increased. In the 2005 dataset a sensitivity test was carried out concerning salt removal efficiency by drainage. Reducing the efficiency of salt removal by drainage had little impact on the results in this case.

5.1.3. HCAB (Copenhagen)

The results for Copenhagen are shown in Figs. 5–8. In Copenhagen an urban background station measuring salt is available and this is subtracted from the traffic site concentrations to give a net concentration. As such negative concentrations are possible and these are kept in the data to show the uncertainty in this net calculation.

Generally, over the four years of data, we see some peaks being reproduced but others not. There is one day in February 2011 (Fig. 6) when a single very large peak of salt is observed. This corresponds to a dry day with salting and may represent the immediate suspension of dry salt that cannot be represented by the model, Section 3.5. Another interesting period is the last week of

Table 2
Type, description and impact of the sensitivity calculations for the 10 datasets. The 'Sensitivity type' refers to the categorisation listed in the text.

Site and period	Sensitivity test 1 description and [type]	Sensitivity test 2 description and [type]
Hornsgatan 2012	No dust binding [3]	Observed surface moisture used for surface retention instead of modelled moisture [3]
Hornsgatan 2013	No dust binding [3]	No snow 18–20 March [1]
RV4 2004	Vehicle spray rate halved reducing the removal of water and salt [3]	Spray rate doubled increasing the removal of water and salt [3]
RV4 2005	Vehicle spray rate halved reducing the removal of water and salt [3]	Drainage efficiency of salt removal reduced from 1.0 to 0.1 leading to less salt removal [3]
Hämeentie 2014	No precipitation on 20 March [2]	Drainage efficiency of salt removal from 1.0 to 0.1 leading to less salt removal [3]
Kehä III 2013	Vehicle spray rate halved reducing the removal of water and salt [3]	No speed dependence for suspension, in this case leading to lower suspension rates [4]
HCAB 2010	Doubled the salt and dust retention threshold ($g_{retention\text{-}min}$) leading to 'drier' surfaces [3]	No speed dependence for suspension, in this case leading to higher suspension rates [4]
HCAB 2011	No salting on 23 February [2]	Additional snow 23 February (1 mm) [1]
HCAB 2012	No precipitation 29 January (1 mm) [1]	Double salt mass application for the whole period [2]
HCAB 2013	Alternative precipitation from Botanic Gardens on 4 critical days (Feb 9, March 19, 20, 28 and 29) [1]	Drainage cut off threshold reduced from 0.5 mm to 0.25 mm leading to more drainage removal of water and salt [3]

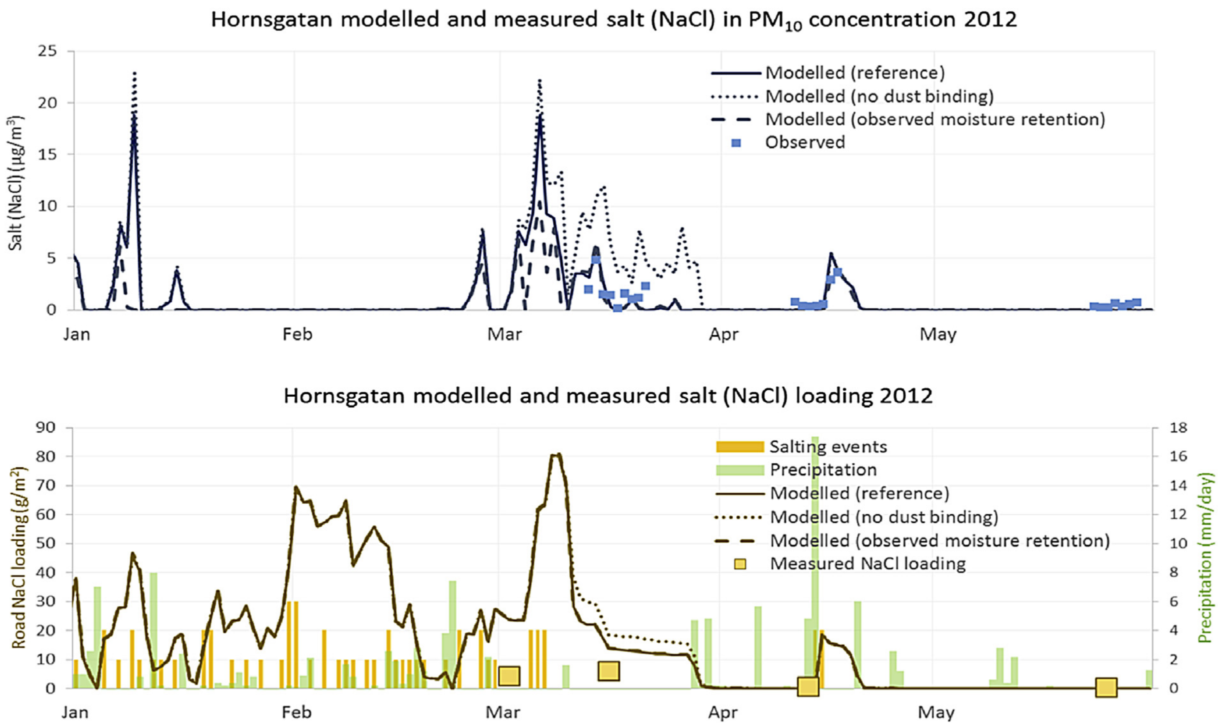

Fig. 1. Modelled and measured daily mean ambient air PM_{10} salt concentrations (top) and road surface salt loading (bottom) for the period January to May 2012 at the Hornsgatan site. Also shown in the lower plot are the salting application events (g/m^2/day) and the precipitation events (mm/day).

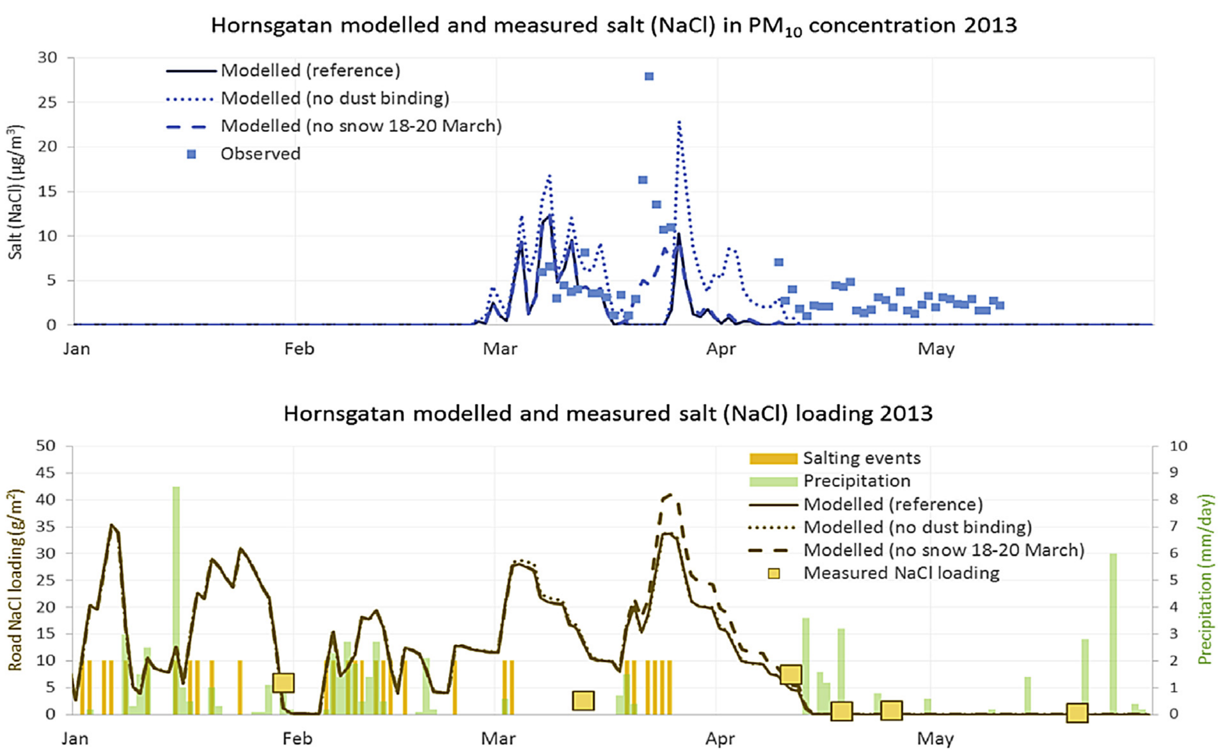

Fig. 2. As in Fig. 1 but for the period January to May 2013.

February 2010 (Fig. 5) where salt contributions to PM_{10} of 10 µg/m^3 are observed whilst the model predicts a wet road without any emissions. This period is characterized by significant precipitation and the road should indeed be wet. It is not clear where this salt contribution comes from.

The mismatch of model with observations in March–April 2011 (Fig. 6) is interesting as it is the result of three salting events on 22 and 23 of February. It is not clear why these salting operations were

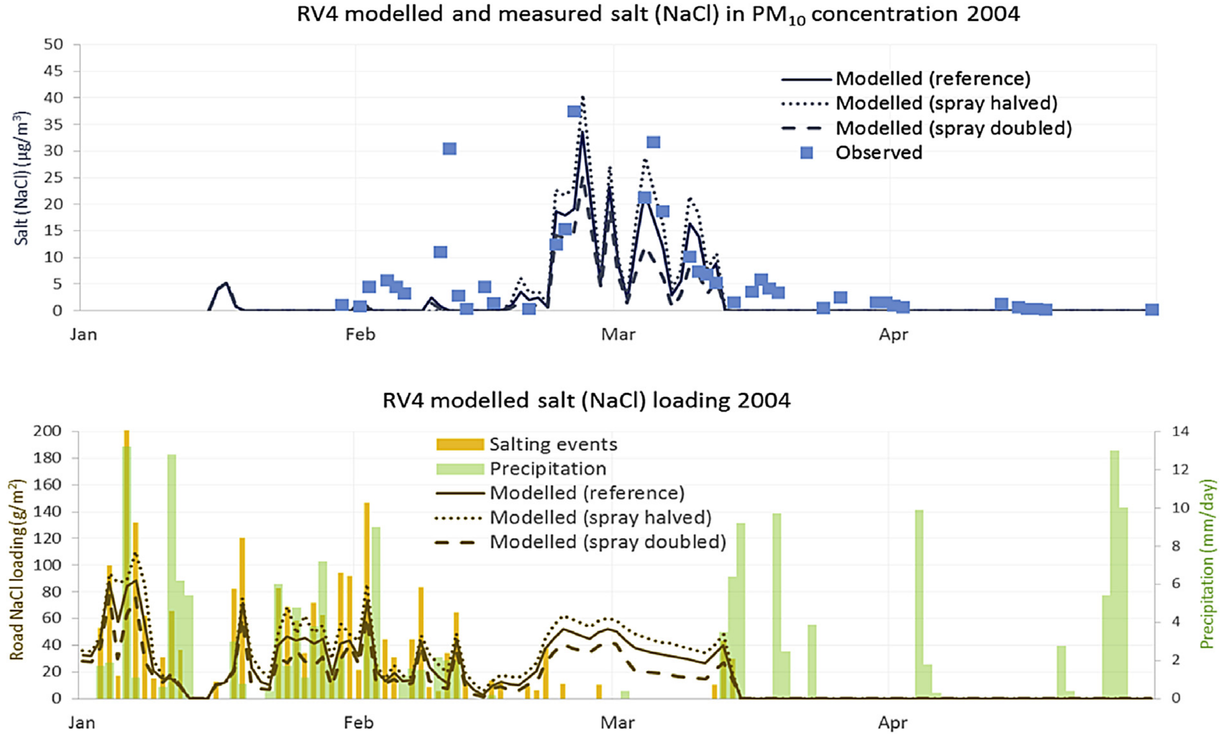

Fig. 3. Modelled and measured daily mean ambient air PM_{10} salt concentrations (top) and modelled road surface salt loading (bottom) for the period January 2004 to April 2004 at the RV4 site. Also shown in the lower plot are the salting application events ($g/m^2/day$) and the precipitation events (mm/day).

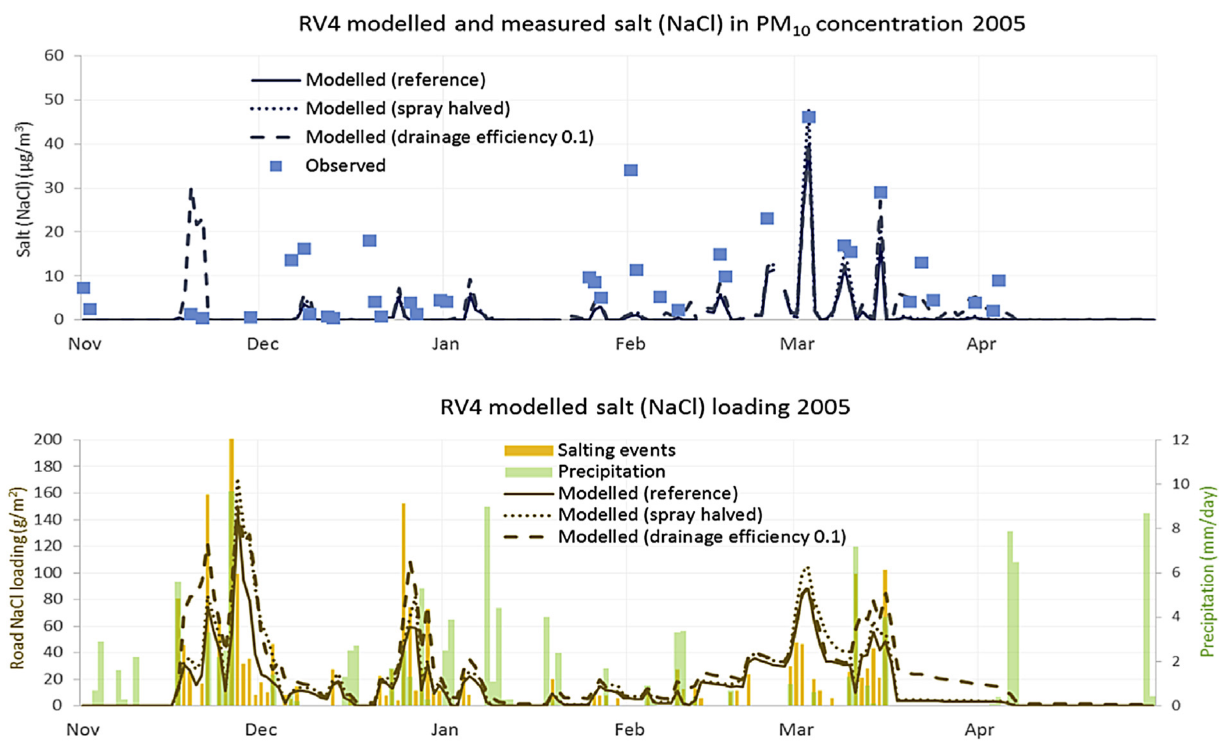

Fig. 4. As in Fig. 3 but for the period November 2004 to April 2005.

carried out as the period was dry as indicated by the precipitation measurements. Salting, especially twice in one day, is usually initiated by precipitation events. If precipitation did occur during these salting operations then the model levels would be significantly lower for this period. We assess the model sensitivity to this one day by removing the salting events on the 23 Feb and by adding 1 mm of snow on this day. Both these changes result in significant differences in the time series.

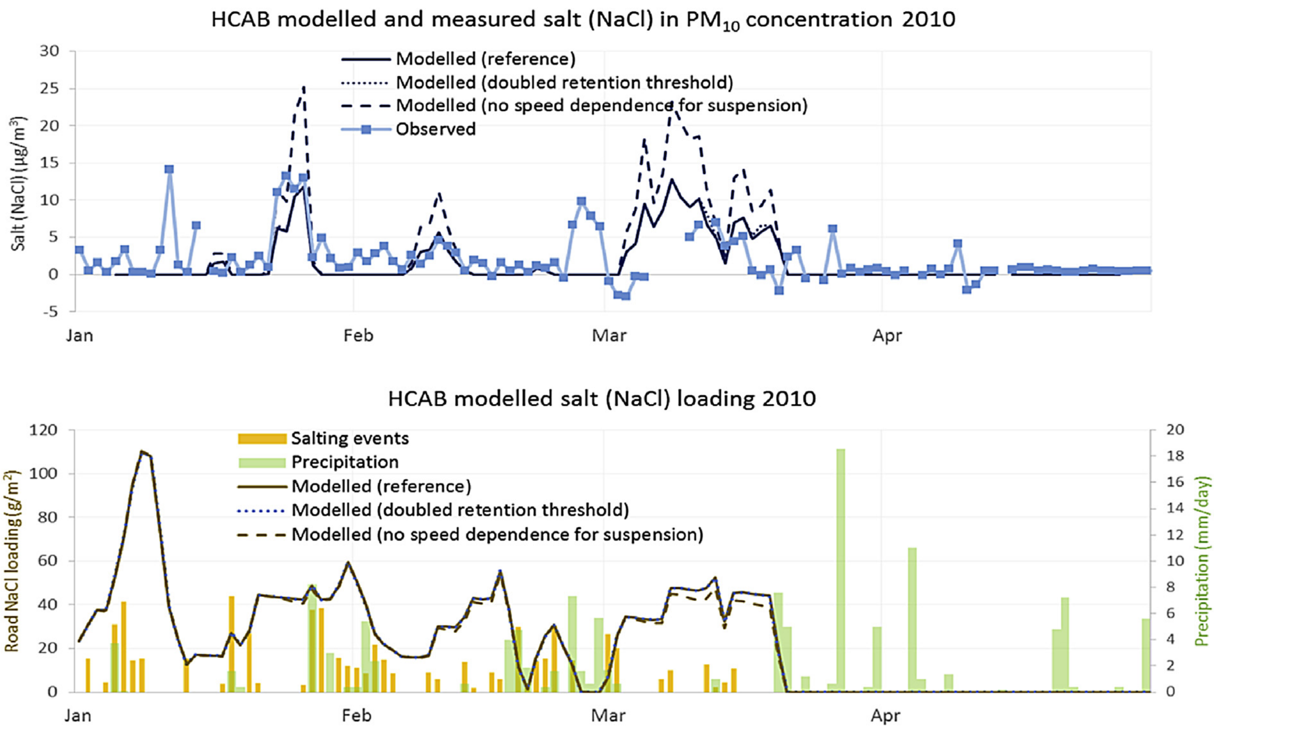

Fig. 5. Modelled and measured daily mean ambient air PM_{10} salt concentrations (top) and modelled road surface salt loading (bottom) for the period January 2010 to April 2010 at the HCAB site. Also shown in the lower plot are the salting application events (g/m^2/day) and the precipitation events (mm/day).

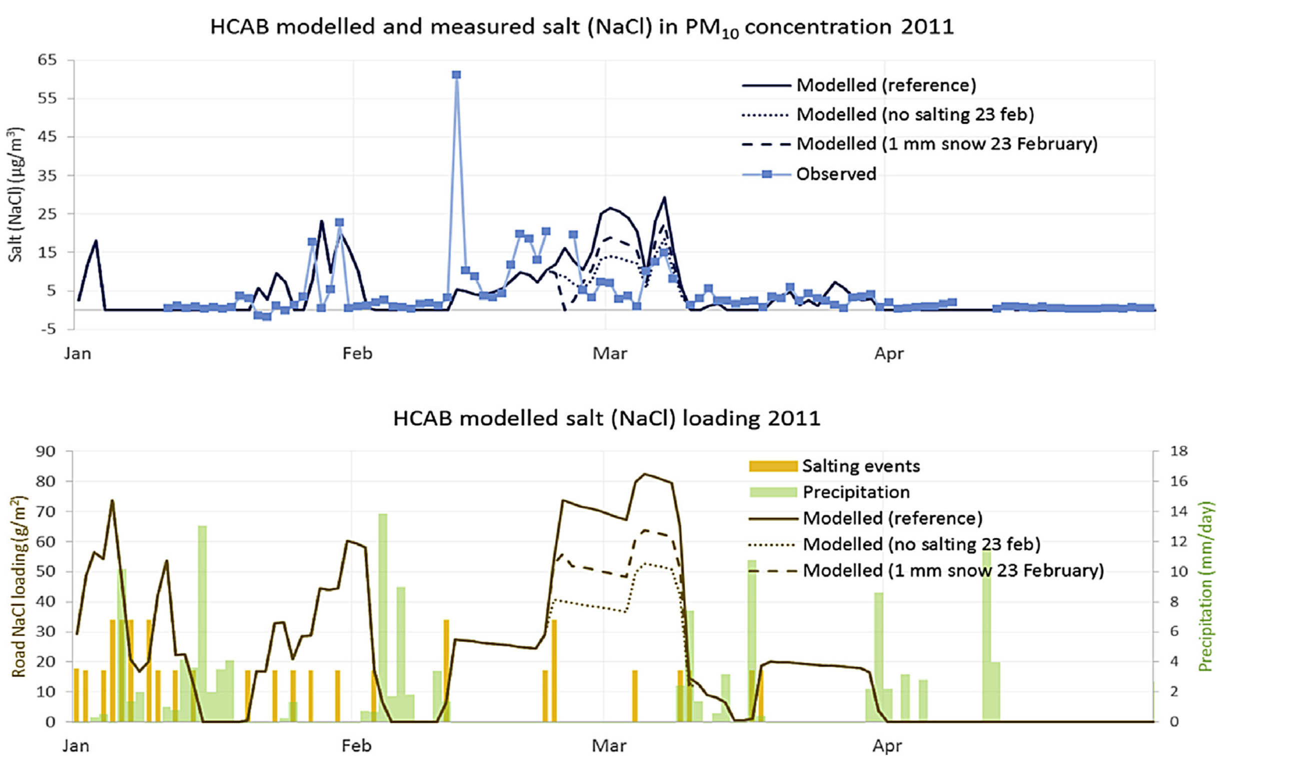

Fig. 6. As in Fig. 5 but for the period January 2011 to April 2011.

At the start of February 2012 (Fig. 7) significant salt concentrations were measured. These results are not reproduced by the model. The highest peak in the modelled salt occurred several days later and was less than half the intensity. To see if this was a result of input data the small precipitation event on 29 January 2012 was removed. This led to some increase in the salt emissions during this peak period but was not sufficient to reproduce the measured concentrations.

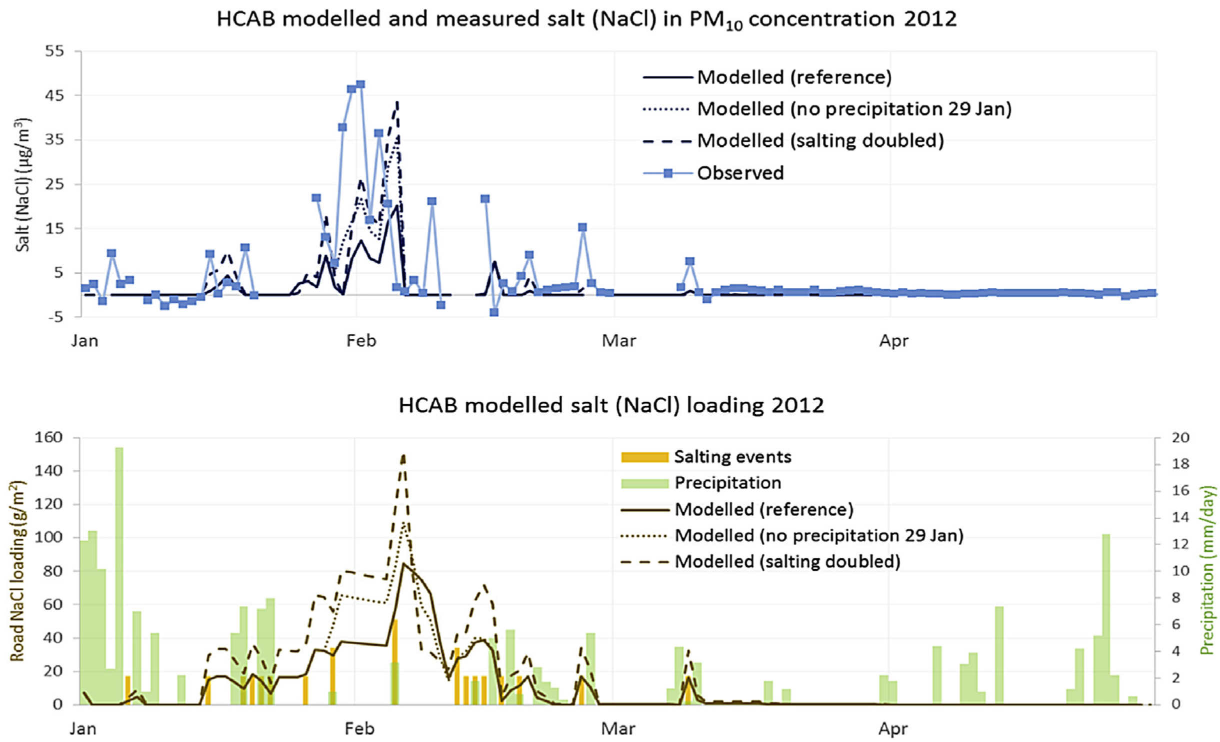

Fig. 7. As in Fig. 5 but for the period January 2012 to April 2012.

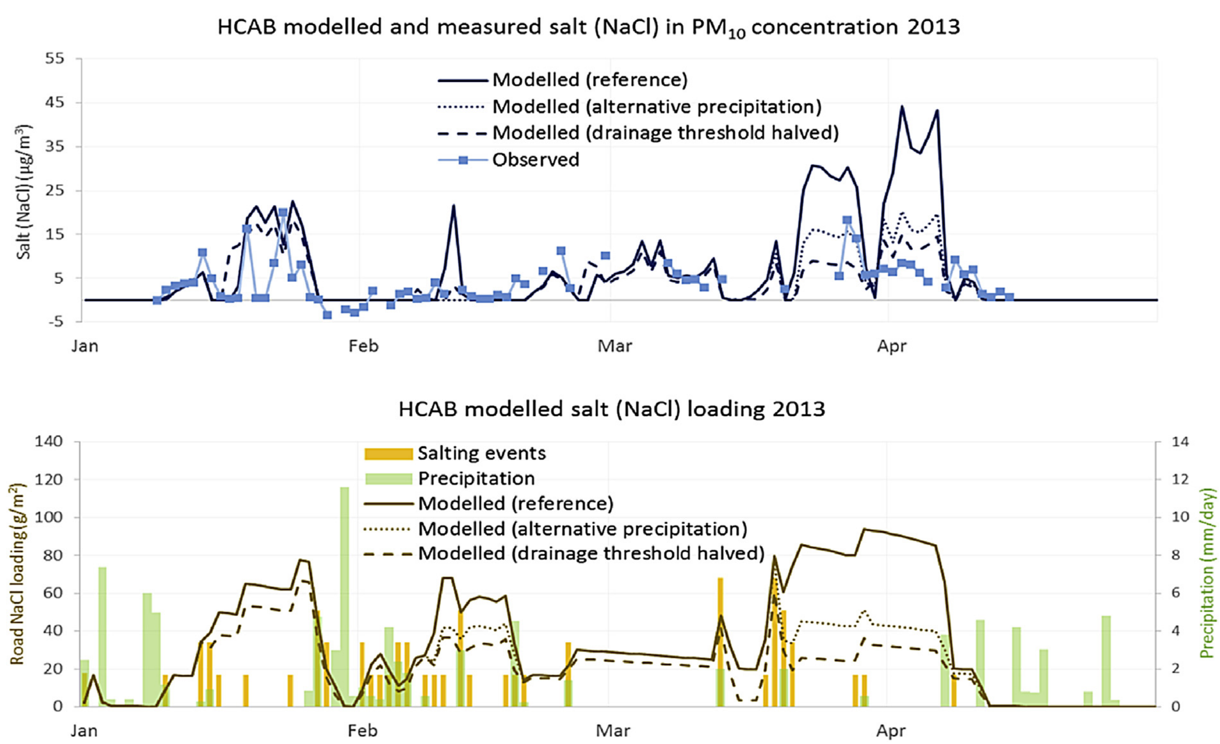

Fig. 8. As in Fig. 5 but for the period January 2013 to April 2013.

In 2013 (Fig. 8) the reference model calculation performs poorly in the period at the end of March to April. Investigation of the precipitation data, for which two sets are available, showed that precipitation was registered at the non-reference site at the end of March 2013 that was not present in the reference precipitation data. As a result, the two precipitation datasets were compared and these were changed when significant differences were seen. The sensitivity run using the alternative precipitation shows a much closer correspondence to the observed salt emissions.

5.1.4. Hämeentie (Helsinki metropolitan area)

For Hämeentie two regional background salt measurements (Utö and Virolahti, located roughly 200 km to the East and West of Helsinki) have been averaged and subtracted from the local measurements for the comparison. Based on this it was found that roughly one third of the measured salt concentrations at Hämeentie are due to sea salt. Results are shown in Fig. 9 where it can be seen that the model correctly predicts the observed salt concentrations in mid-March but fails to reproduce the observed salt for the rest of the period. Two weeks prior to the mid-March salting only one salting event occurred (1 March) before significant precipitation events (3 March), so there was no modelled salt available for suspension. After the mid-March salt emission event 6 mm of snowfall (20 March) removed salt from the surface through drainage, yet measurements indicate enhanced levels of ambient salt for the week following this precipitation. Sensitivity tests with salt drainage efficiency and by removing this precipitation event showed that salt could persist over longer periods.

5.1.5. Kehä III (Helsinki metropolitan area)

The NaCl measurement results on Kehä III are limited to a one month dry period in March–April 2013, Fig. 10. In this case the model successfully captures the observed salt level trends. Reduction of the suspension rate, by removing the speed dependency of the suspension so that it is valid for the reference speed of 50 km/h reduces the modelled concentrations. Reducing the spray removal increases the concentrations, as for RV4 in Fig. 3.

5.2. Statistical summary

Figs. 11 and 12 provide a statistical summary of the results for all sites and periods presented in Figs. 1–10. Of the ten reference calculations shown six are within a factor of two of the observed mean concentrations. The observed mean concentration, over all the datasets, is 4.2 μg/m^3 and the modelled mean is 2.8 μg/m^3, giving a fractional bias of −0.38. The RMSE of the mean concentrations, over all the datasets, is 2.9 μg/m^3. HCAB 2012 shows the poorest result in terms of bias.

In Fig. 12 the coefficient of determination (R^2) is shown for each of the datasets. Some datasets are highly correlated, e.g. RV4 datasets, whilst others, e.g. Hornsgatan 2013 reference calculation, are not correlated at all. Only the sensitivity run with the removal of the snow events in March on Hornsgatan 2013 changes the correlation significantly. For most datasets the sensitivity tests did not change the correlation to a significant degree. The average R^2 for all the datasets is found to be 0.28. T-tests were applied for each dataset to assess the significance of the results. P-values < 0.05 were found in all cases so the R^2 determined are interpreted as being significant.

5.3. Sensitivity analysis summary

In Table 2 a description of the various sensitivity studies carried out for the 10 datasets is provided. Though some of the sensitivity tests were commented in Section 5.1 we summarise the results of the sensitivity studies based on the four sensitivity types defined in Section 4.

1. Precipitation: The model can be quite sensitive to individual precipitation events. This is clearly shown in the HCAB 2011–2013 datasets (Figs. 6–8) where alternative precipitation input data can lead to significant changes in the time series as well as the mean concentrations. For example removing 1 mm of snowfall on 29 January 2012 on HCAB leads to a 70% increase in the mean modelled salt concentration for the whole period (Fig. 11).

2. Salt input: In all cases salting was logged to some degree, however this was usually only specified to occur within a particular time interval and within a particular region or road category. Generally the amount of salt used was also not specified so

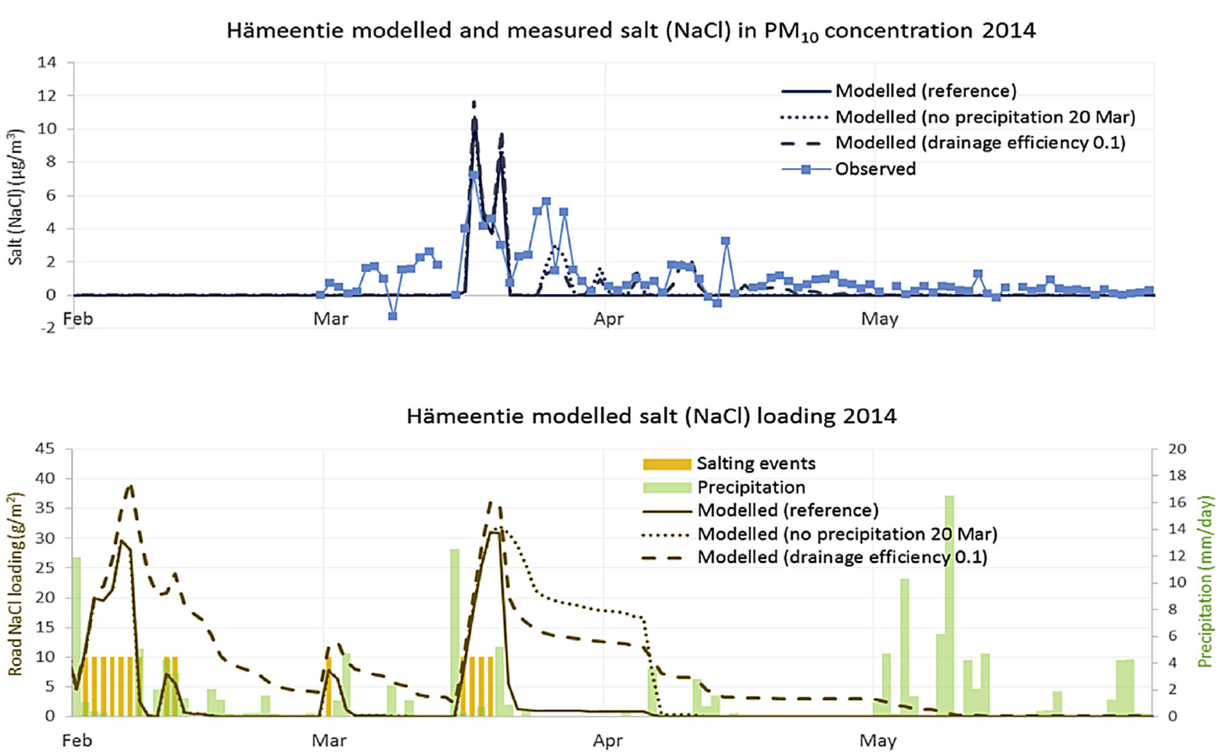

Fig. 9. Modelled and measured daily mean ambient air PM_{10} salt concentrations (top) and modelled road surface salt loading (bottom) for the period February 2014 to May 2014 at the Hämeentie site. Also shown in the lower plot are the salting application events (g/m^2/day) and the precipitation events (mm/day).

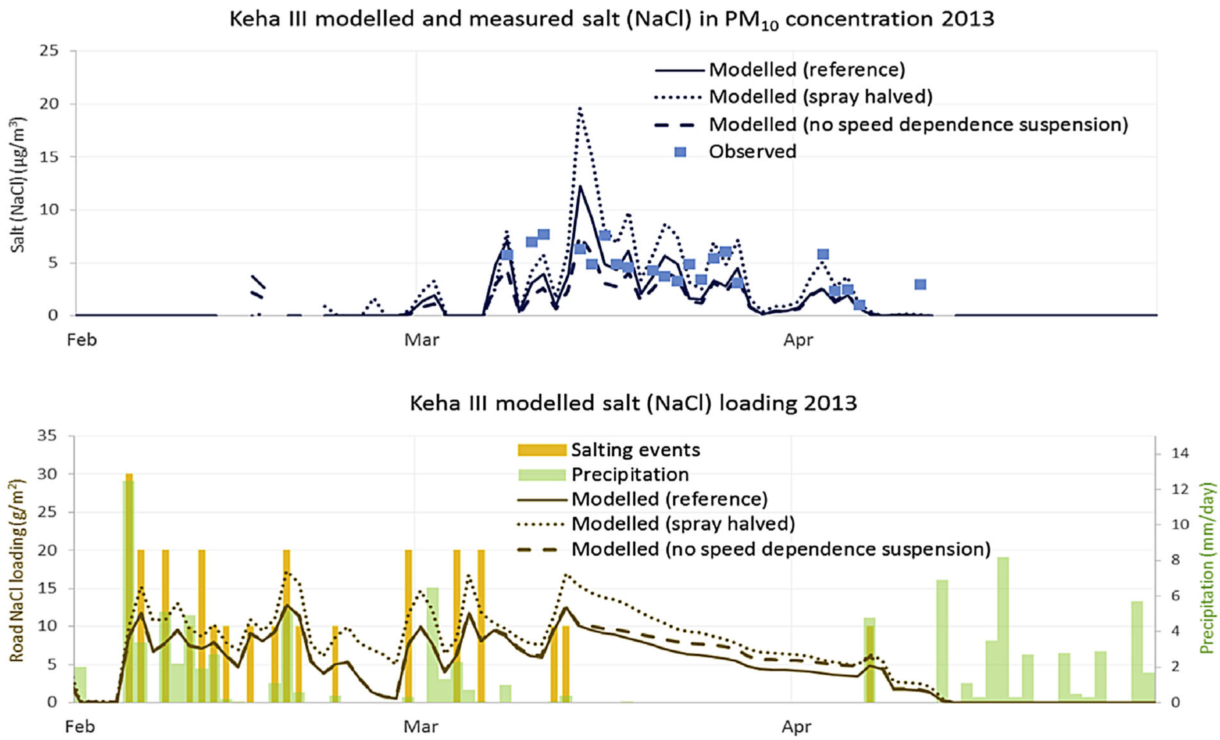

Fig. 10. Modelled and measured daily mean ambient air PM_{10} salt concentrations (top) and modelled road surface salt loading (bottom) for the period February to April 2013 at the Kehä III site. Also shown in the lower plot are the salting application events (g/m^2/day) and the precipitation events (mm/day).

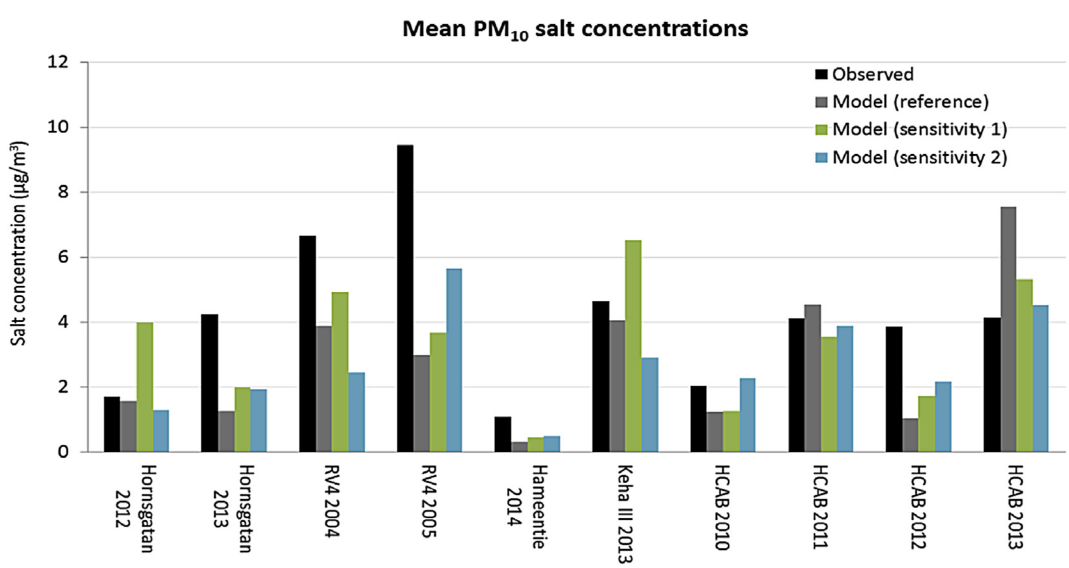

Fig. 11. Mean observed and modelled concentrations for the 10 datasets including reference and sensitivity calculations, see Table 2.

assumed average salt masses were used. There is significant uncertainty as to when salting took place and how much salt was applied. In one case (HCAB, 2011) one days salting was removed from the model input. This had a visible impact on the time series (Fig. 6) but less on the mean and correlation. One dataset, HCAB 2012 (Fig. 12), was tested by doubling the total salt and this led to a 110% increase in the mean salt concentrations.

3. *Moisture parameters:* Of the model parameters tested the most sensitive was the inclusion, or not, of dust binding in the model (Hornsgatan, 2012, 2013, Figs. 1–2). The model was also sensitive to the vehicle spray rate, RV4 (Figs. 3–4) and Kehä III (Fig. 10), with a significant increase in salt emissions as a result of reduced spray. These two roads have high vehicle speeds and the salt budget (Fig. 13) shows that spray is the most important removal term for salt for these two roads. For most roads drainage is the major sink term for salt and the sensitivity tests concerning drainage, either threshold, HCAB 2013 (Fig. 8), or efficiency of salt removal, RV4 2005 (Fig. 4) and Hämeentie 2014 (Fig. 9), show that salt emissions are also sensitive to these parameters.

4. *Suspension parameters:* Sensitivity of salt emissions to suspension was assessed for Kehä III 2013 (Fig. 4) and HCAB 2010 (Fig. 5). In both cases the speed dependence of suspension was

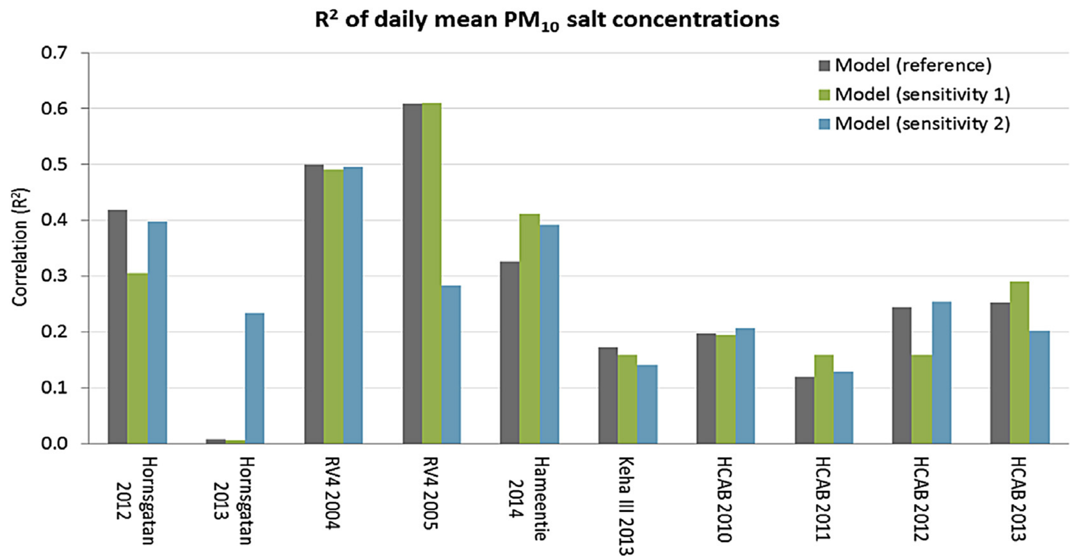

Fig. 12. R^2 of the observed and modelled concentrations for the 10 datasets including reference and sensitivity calculations, see Table 2.

removed so that the suspension rate applied was valid for the reference speed of 50 km/h. This led to an 85% increase in the suspension for HCAB (average speed of 29 km/h) and a reduction of 29% for Kehä III (average speed 83 km/h).

In general we see that the model is strongly sensitive to some input data. In particular precipitation has a strong impact on the emissions, however it is clear that salting activities, both timing and amount, are also important input data. The model is also sensitive to a number of the model parameters. Parameters related to the major sinks of the salt budget (drainage and spray) show the most sensitivity.

5.4. Salt loading budget and suspension

For each of the reference model calculations a salt loading budget has been determined. This budget represents the accumulated salt production and removal over the modelling period for each of the ten datasets and is shown in Fig. 13. The amount of salt applied varies from site to site with the largest salt application found at the Oslo site RV4 and the least salt applied at the Helsinki site Hämeentie. The major removal term is drainage but for roads with high speed signage (Kehä III and RV4) vehicle spray is also an important removal term. This reflects measurements made of near road deposition of salt from splash and spray processes, e.g. Blomqvist and Johansson (1999), which show that spray is a major removal process on high speed roads. The removal by suspension is a very small term in the budget. On average, for all the sites, suspension removal accounts for just 2% of the total salt removal.

Since only 28% of the modelled suspended salt is in the PM_{10} fraction, also shown in Fig. 13, then on average just 0.5% of the applied salt is emitted as PM_{10}. Though this fraction clearly varies from site to site and period to period it is worth providing a general estimate of the contribution of applied salt to kerbside PM_{10} concentrations. Based on the observed and modelled calculations we find an average increase in kerbside winter mean salt concentrations of 4.1 $\mu g/m^3$ for every kg/m^2 of salt applied. The RMSE of this

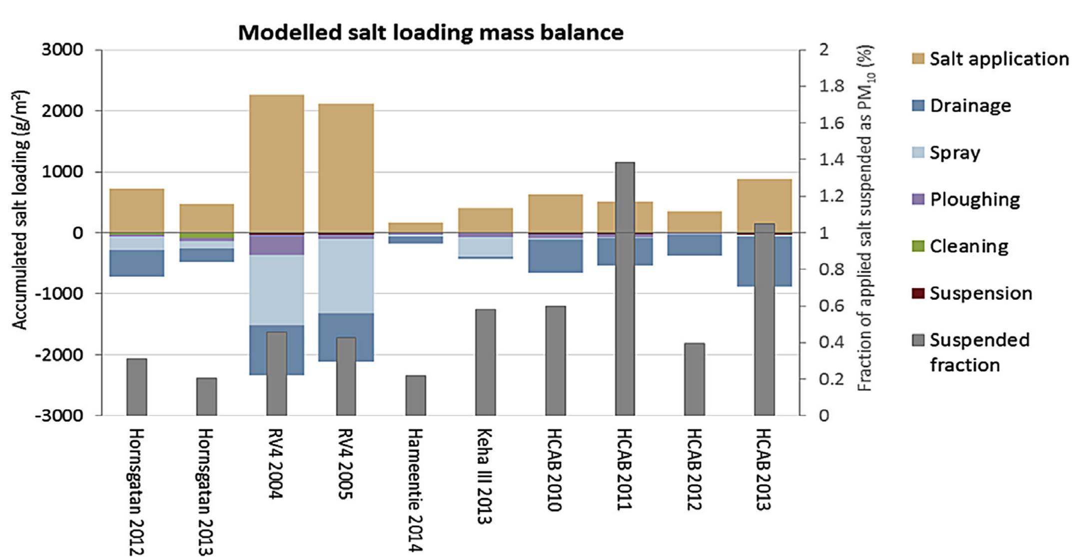

Fig. 13. Modelled accumulated salt mass balance for the ten datasets studied. Also shown is the suspended fraction of the total applied salt in PM_{10} (grey bars).

estimate, due to model uncertainty and natural variability from site to site, is determined to be 3.4 μg/m^3 for every kg/m^2 of salt applied.

6. Conclusion and discussion

In this paper we present the first modelling study of road salt emissions. Included are ten new datasets of observed salt concentrations in ambient air that also includes information concerning salt application. The major aim of this study is to assess the performance of the NORTRIP model in predicting emissions of road salt. Of the ten observational datasets assessed two of these (Hornsgatan, 2012 and RV4 2004) show a good agreement between modelled and observed concentrations in regard to both mean concentration (less than a factor of 2) and correlation ($R^2 > 0.4$). Four other modelled datasets (HCAB, 2010; HCAB, 2011; HCAB, 2013 and Kehä III, 2013) show mean concentrations within a factor of 2 of the observations but have an R^2 between 0.1 and 0.3. One dataset (RV4 2005) shows a high correlation ($R^2 = 0.6$) but underestimates the concentrations by a factor of 3. The remaining three dataset calculations (Hornsgatan, 2013; Hämeentie, 2014 and HCAB, 2012) perform less well, Figs. 11 and 12.

The observed mean concentration, over all ten datasets, is 4.2 μg/m^3 and the modelled mean is 2.8 μg/m^3, giving a fractional bias of −0.38. The RMSE over all the datasets is 2.9 μg/m^3. In four of the datasets (HCAB) urban background salt concentrations were subtracted for comparison with the model. For one dataset (Hämeentie) regional background concentrations were subtracted. It is thus expected that the model would underestimate the observed concentrations in all cases except HCAB. The average PM_{10} concentration of road salt, calculated by the model for all 10 datasets, is slightly larger than the calculated exhaust contribution (2.6 μg/m^3). This indicates the importance of salt as a traffic related source of PM_{10}.

An assessment of the modelled salt mass balance indicates that the major removal process for salt is drainage, but on roads with high speed signage vehicle spray can be more important. The model calculations indicate that on average only 0.5% of the applied salt is suspended in the PM_{10} size fraction, indicating that suspension is generally not a major contributor to the surface mass balance. Based on the observed and modelled calculations for these datasets we estimate that kerbside winter mean salt concentrations increase by 4.1 ± 3.4 μg/m^3 for every kg/m^2 of salt applied during the winter season.

Modelling of road salt emissions is a demanding task, requiring the model to correctly predict the road salt loading, the road surface conditions and the road salt suspension. This in turn requires the correct mass balance for salt, including the production by salt application and removal by drainage and vehicle spray. For the surface conditions the correct mass balance of the surface water/ice/snow is required, involving the same wet removal processes as for salt but also including evaporation and condensation through energy balance considerations. Important input data is then the salt application information and the meteorological data, particularly precipitation. If these input data are not correct then the results of the modelling are also not correct.

Beyond the uncertainties in the input data there are a range of model parameterisations that are uncertain and for which the model is sensitive. In order to explore some of these uncertainties 20 additional sensitivity studies were carried out using the ten datasets. These helped to indicate the range of model results based on uncertainties in the input data and in a number of model parameters. The results were sensitive to model parameters that affected the surface salt mass balance and suspension, but were also sensitive to input data such as precipitation and salt application. The total uncertainty in the mean concentrations is indicated by the mean RMSE of 2.9 μg/m^3, which is as large as the model mean concentrations of 2.8 μg/m^3. This is the combined result of model uncertainty, input data uncertainty and measurement representativeness uncertainty (only local road contributions or including urban and regional background). Since we cannot separate these uncertainty sources we then estimate the uncertainty of the modelled salt concentrations to be around a factor of 2, assuming that reasonable input data is available.

From this study it is clear that input data, particularly salt application and precipitation, is particularly important for correctly predicting salt emissions and for further development of the model. For future comparisons more controlled data and conditions are required. Future field experiments should include both kerbside and urban background ambient air PM_{10} salt concentrations, regular salt loading measurements from the road surface, drainage measurements, spray measurements, surface wetness measurements and accurate salt application data. This would provide more consistent data for model evaluation and for the further improvement of salt process descriptions.

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