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Sensitivity studies

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EMECAP Deliverable 5.3

1 Introduction

As part of Work Package 5 of the EMECAP project, dispersion model calculations have been carried out in order to determine the concentration and deposition of Mercury in the region surrounding the selected MCCA plants. The 3 plants under study are the Bohus plant in Sweden, the Rosignano Solvay plant in Italy and the Tarnow plant in Poland. Measurement campaigns at all 3 plants during the EMECAP project have provided essential data to allow validation and verification of dispersion model results.

The dispersion and deposition of Mercury around MCCA plants is a three fold problem. The first is the dispersion itself, which is dependant on local meteorology and emissions from the plant. The second is the Mercury chemistry and the third is the deposition rate for varying Mercury species. Mercury can be found in several forms as a result of chemical reactions with other gases, in particular Chlorine, to transform Gaseous Elemental Mercury (GEM) into the more reactive species, Reactive Gaseous Mercury (RGM). The sum of these is known as the Total Gaseous Mercury (TGM). In addition Mercury can deposit on particles, Total Particulate Mercury (TPM). The deposition rates of these different forms can differ by up to 2 orders of magnitude, with RGM expected to have the highest dry and wet depositions.

Results from the previous report, D5.2, have shown that chemical reactions inside the plant are a likely source for the formation of RGM, which is the Mercury species most easily deposited to the surface. Chemical reactions in the atmosphere, on short time scales, do not appear to play an important role in the formation of RGM as the dispersion of pollutants quickly reduces their concentration. The most likely site for the formation of RGM is thus in the factory itself.

Mercury can be deposited through dry deposition processes or through wash out of Mercury during precipitation events, wet deposition. The dry deposition velocities and wet deposition scavenging rates are not well defined for any of the Mercury species. In order to study the sensitivity of Mercury deposition in the local region surrounding the MCCA plants a number of sensitivity tests are carried out to determine local deposition around the plant as a function of dry deposition velocities, wet scavenging ratios and the percentage of Mercury emitted in the form of RGM. Sensitivity runs have been carried out at the Bohus plant in Sweden 5 day periods.

The results indicate that uncertainty in the deposition parameters, particularly the wet scavenging ratio, is significant in determining the total deposition. They also show that RGM is the most important species contributing to deposition despite its low representation, < 1% of TGM.

2 Description of the models

Two models are employed in the current study. The first is 'The Atmospheric Pollution Model' (TAPM) from CSIRO in Australia, which is used for meteorological calculations. The second model is an off-line dispersion chemistry model called EPISODE. This model has been adapted to include a Mercury-Chlorine chemistry scheme and is used to calculate deposition and concentration fields of GEM, RGM and TPM.

2.1 The meteorology and dispersion model TAPM

TAPM has been developed by CSIRO in Australia as a complete pollution model that includes meteorology, dispersion and a limited photochemistry scheme (Hurley, 2002). The heart of TAPM is the meteorological model. This can be nested into a regional scale model, in this case the LAPS model (Puri et al. 1998), starting at a resolution of 10's of kilometres and reducing down with each nest to a grid spacing of around 1 km. In the current study 4 nestings have been implemented down to a resolution of 500m. Within TAPM are worldwide land-use and sea surface temperature data sets that can be used for surface exchange calculations. In addition to the boundary conditions set by the regional scale model, input of local wind measurements can be used to 'nudge' the local wind field towards local observations.

In addition to meteorology, TAPM can also calculate the transport and dispersion of pollutants on a pollution grid. However, this scheme is not suitable for the current studies and so a second dispersion model has been implemented, the EPISODE model.

2.2 The chemistry/transport model EPISODE

In order to calculate Mercury chemistry and deposition an off-line model, using TAPM meteorology fields, has been used. This is the EPISODE model (Slørdal et al., 2003), especially adapted to calculate Mercury chemistry and deposition. It consists of a transport and dispersion scheme, similar to TAPM, and a Mercury/Chlorine/Ozone chemistry scheme. Dry deposition is calculated by using predefined deposition velocities and wet deposition occurs during precipitation events using a wet scavenging parameterisation. In the EMECAP report D5.2 it was shown that atmospheric chemistry was not important for the transformation of Mercury species and so no chemistry has been included in these model runs.

2.3 Dry deposition

Dry deposition within the EPISODE model is calculated using fixed deposition velocities (V_d) with the formulation

$$Dry\ Deposition\ flux = V_d\, C$$

where C is the concentration of the species near the surface. In this case the lowest model level, which is at 10 m.

The deposition velocities used in this study have been estimated from the currently available data in the literature. Below is a list of dry deposition velocities used in various models or determined by observations.

The dry deposition velocity for all Mercury species is not well defined. Observations made in forests (Rea et al., 2002) indicate higher deposition velocities for GEM than for soils (Ilyin et al., 2002), but this calculation has been made assuming that GEM was the only form of Mercury present. A summary of dry deposition velocities for GEM can be found in Ilyin et al. (2002) and the range is quite large, from 0 to 0.1 cm/s. It is most likely that the dry deposition velocity of GEM is low and a value of 0.02 cm/s is assumed as the default for model runs in this study.

Dry deposition velocities for RGM are always taken to be high and often made equal, in model calculations, to the dry deposition velocity of Nitric acid, even though there is no experimental evidence to support this. For the current study a value of 2.0 cm/s is assumed.

Table 1. Summary of dry deposition velocities available in the literature. References:(2) Rea et al., (2002), (3) Landis et al. (2003), (4) Ilyin et al. (2001), (5) Ilyin et al. (2002), (6) Bloxam (1996), (7) Berg et al. (2001)

Dry deposition velocity, V_d, (cm s^{-1})			**Reference**	**Comment**
GEM	**RGM**	**TPM**		
0.05			(2)	Measurements in forest
0.012 - 0.025	1.2 – 2.5		(3)	Used in model calculations
0.01 – 0.03		> 0.05	(4)	Used in EMEP model
0 – 0.1			(5)	Review of models and measurements
0.03	2.0		(6)	Used in ADOM model
0.0	4.0	0.2	(7)	Used in HMET model
0.02	2.0	0.1	This study	Default values for sensitivity tests

Similarly, there is no experimental evidence to support dry deposition velocities for TPM. It is often assumed to deposit at the same rate as Sulphate particles in model calculations. We assume a value of 0.1 cm/s.

2.4 Wet deposition

Wet depostion is carried out in the EPISODE model at each model level using the following formulation

$$\frac{\partial C_{(i,j,k)}}{\partial t} = -C_{(i,j,k)} \frac{W.P}{\Delta z_k}$$

where C is the concentration in the model grid (i,j,k), W is the wet scavenging ratio, P is the precipitation (m/s) and Δz is the depth of the model grid volume. This can be rewritten at every time step as

$$C_{(i,j,k)}(t+\Delta t) = C_{(i,j,k)}(t)\left(\exp(-\frac{W.P}{\Delta z_k}\Delta t) \right)$$

and the total mass of species deposited, *M*, in the period Δt is given by

$$M_{wdep}(\Delta t) = \sum_k C_{(i,j,k)}(t)\left(1 - \exp(-\frac{W.P}{\Delta z_k}\Delta t) \right)\Delta z_k$$

The wet deposition scavenging ratio, *W*, thus defines the efficiency with which the species is removed from the atmosphere during precipitation events.

There is little experimental evidence to determine *W* for Mercury and so it is usually correlated with other species such as Nitric acid and Sulphate, as is done in the EMEP model. Below are two references to wet scavenging ratios used in the literature.

Table 2. Summary of wet scavenging ratios available in the literature. References:(1) Abbott et al., (2002), (4) Ilyin et al. (2001)

Wet deposition scavenging rate, *W* (x 10^6)			**Reference**	**Comment**
GEM	**RGM**	**TPM**		
		0.2	(1)	Best fit between model and observations
0.0	1.4	0.7	(4)	Used in EMEP model
0.0	1.4	0.7	This study	Default values for sensitivity tests

3 Model calculations

3.1 Model set up

The sensitivity runs have been carried out for a 5 day period at the Bohus plant in Sweden from 01.07.2002 to 05.07.2003 inclusive. The model parameters for TAPM are listed in Table 3.

Table 3. Model parameters used in the TAPM simulation

Horizontal grid dimensions	25 x 25
Horizontal grid size	From 15 km to 500 m in 4 nests
Vertical grid dimensions	30
Vertical grid size	Lowest level 10 m, up to 8000 m

A short time period was selected in order to carry out a number of different scenarios. This particular period was chosen as it corresponded to the second EMECAP campaign at Bohus and it was during a fairly rainy period, which would allow the sensitivity of the model to wet scavenging to be tested. The total rainfall calculated by TAPM during this period was 11.86 mm, which is the equivalent of 865 mm/year. This value is not far from the expected annual mean at Bohus though the duration of precipitation, 75% of the simulation period, is larger than the yearly average.

The emissions from the plant during this period are listed in Table 4. These are based on measurements made during the campaigns and represent average emissions. The absolute values of the emissions are perhaps less important for the sensitivity runs as these are usually described in relative terms.

Table 4. Emissions of Mercury species used in the model simulations

Emissions (g hr^{-1}) and *% of TGM*				
GEM	**RGM**		**TPM**	
8.4	0.059	*0.7%*	0.025	*0.3%*

3.2 Concentration and deposition fields

In Figure 1, the mean concentration fields calculated for GEM and RGM are shown for the modelling period. The labelled stations, S1, S2, S3 and S4 correspond to either measurement sites or wet deposition sites used during the EMECAP campaigns. In Figure 2 the dry deposition fields for GEM and the wet deposition fields for RGM are shown.

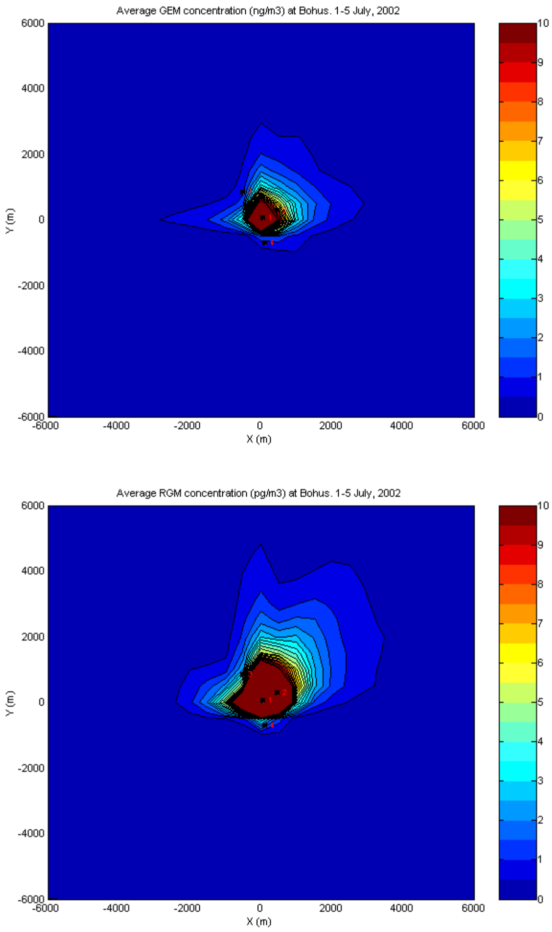
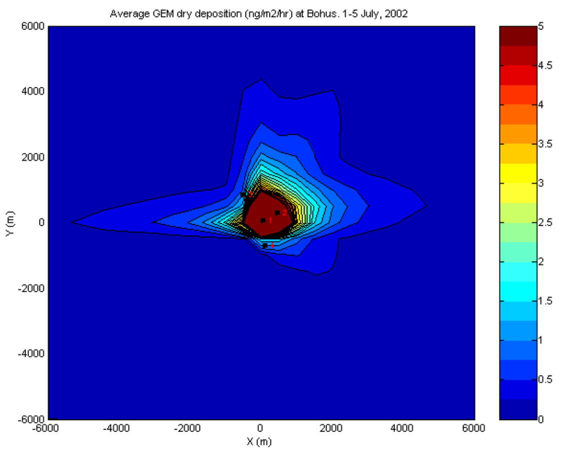


Figure 1. Left: Average concentration of GEM for the 5 day simulation period. Contours are in ng/m^3. Right: Average concentration of RGM for the 5 day simulation period. Contours are in pg/m^3.



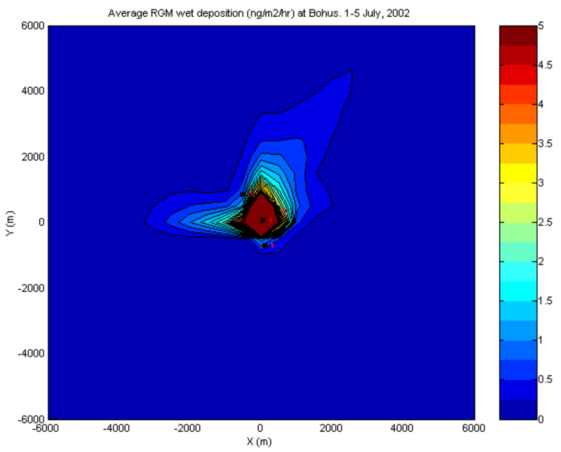


Figure 2. Left: Average dry deposition of GEM for the 5 day simulation period. Contours are in ng/m^2/hr Right: Average wet deposition of RGM for the 5 day simulation period. Contours are in ng/m^2/hr.

The spatial distribution of GEM and RGM is similar but not the same due to the influence of wet deposition processes which alter the spatial distribution for RGM. This is best seen in Figure 2 where the wet deposition field, dependent on precipitation events, shows a different spatial structure to the dry deposition field for GEM. Note that the scale on the deposition fields is the same though the scale on the concentration fields is a factor of 1000 different.

The deposition results are summarized below in Figure 3 and Table 5 where the mean deposition for the 3 species and the two deposition processes are shown as a function of distance from the plant. These values have been calculated by integrating over the model area for the various distances.

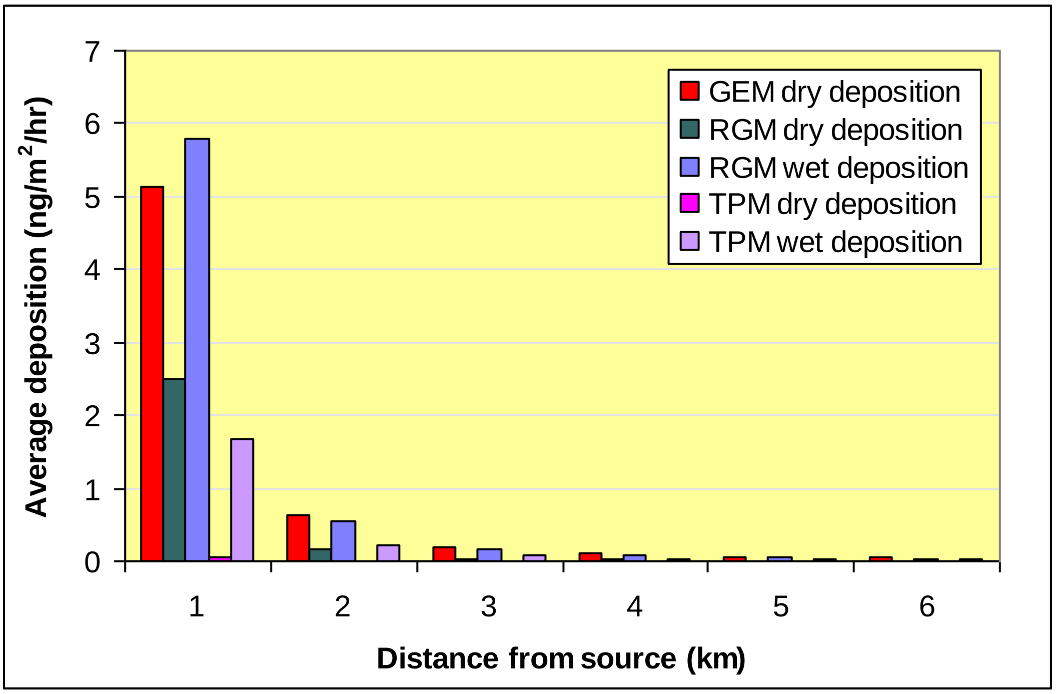


Figure 3. Plot showing the average deposition rates over the 5 day simulation period as a function of distance from the plant. The vertical scale is in ng/m^2/hr.

There are two important points that can be drawn from Figure 3. The first is that the relative contribution of RGM to the deposition of Mercury is similar to that of GEM in spite of the fact that only 0.7% of al the Mercury emitted from the plant is in the form of RGM. This is the result of wet deposition and higher dry deposition rates. The contribution of TPM wet deposition is also significant though dry deposition is not. The second point is that most of the deposition occurs within 1 km of the plant. Around 80% of the RGM that is deposited within a 5 km radius is actually deposited within the first 1 km and 43% of GEM is deposited in this region.

Table 5. Summary of the calculated depositions for the 3 species and 2 deposition processes.

Species and deposition	GEM dry	RGM dry	RGM wet	TPM dry	TPM wet	TGM
Emission of species (g/hr)	8.4	0.059	0.059	0.025	0.025	8.5
Species deposition within a 5 km radius (g/hr)	0.0238	0.0083	0.0234	0.0002	0.008	0.063
Species deposition within a 5 km radius (% of species emission)	0.28 %	14.0 %	39 %	1 %	36 %	0.74 %
% of species deposition that occurs within 1 km	43 %	90 %	75 %	58 %	45 %	60 %

The percentage contribution to deposition is also shown as a function of distance in figure 4. RGM, both dry and wet deposited, is most quickly deposited in the immediate vicinity of the plant due to the quick reduction in RGM concentrations by both deposition and dispersion. GEM concentrations are reduced almost only by dispersion and as such do not reduce as rapidly.

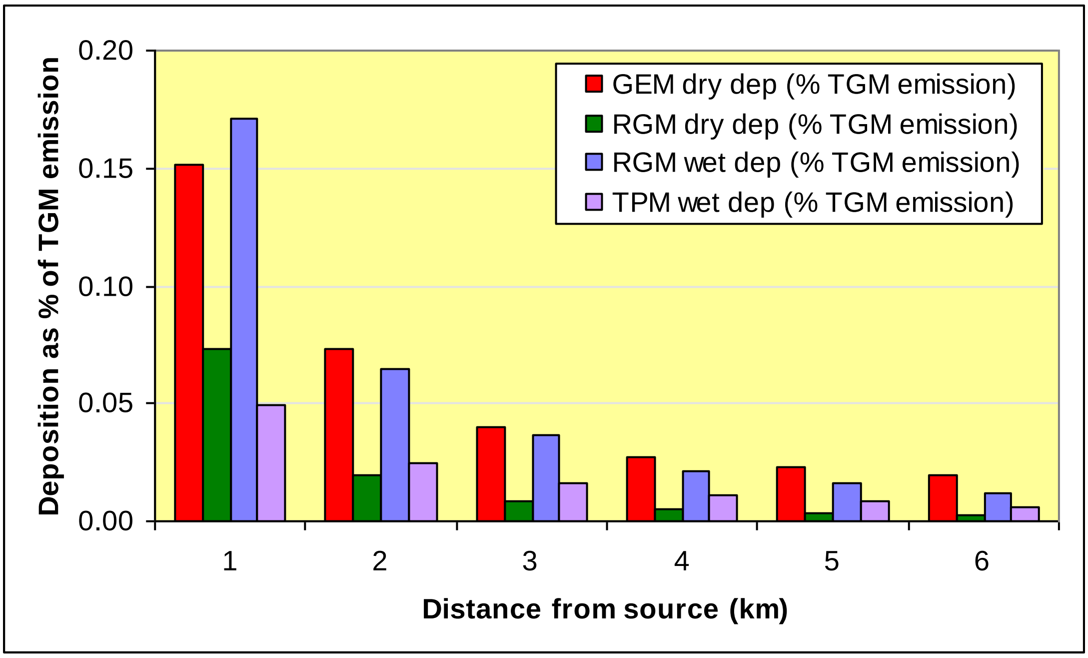


Figure 4. Plot showing deposition as % emitted TGM, averaged over the 5 day simulation period, as a function of distance from the plant.

Due to the higher deposition rate of RGM the relative concentrations of RGM/GEM decreases with distance from the plant. This is shown in figure 5 for two scenarios, one in which no wet deposition takes place and the other in which no dry deposition occurs.

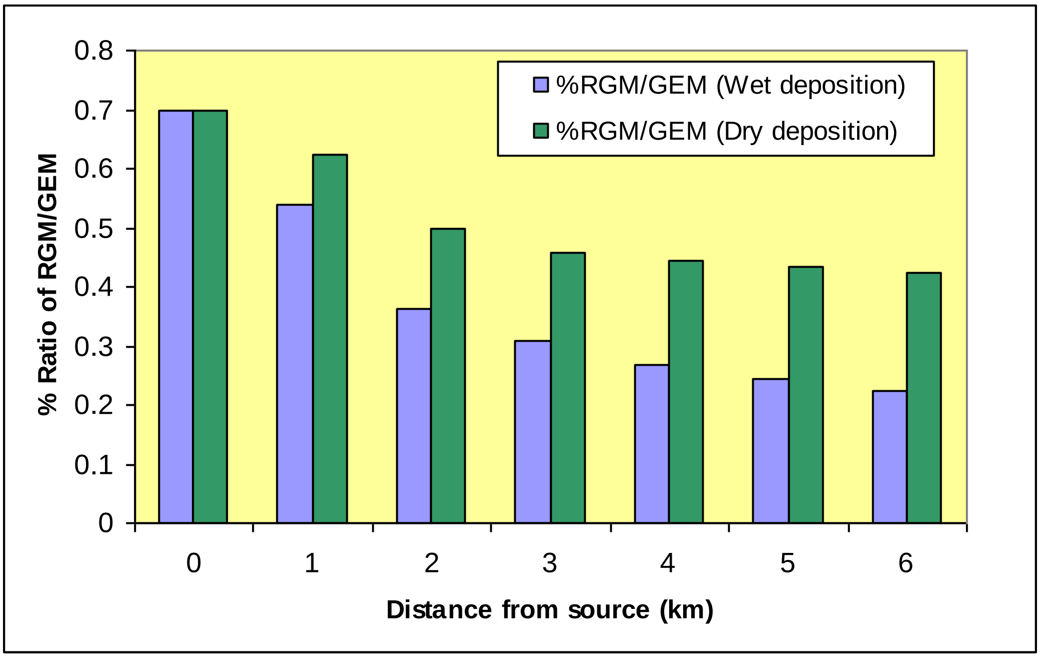


Figure 5. Plot showing the relative concentration of RGM to GEM in % as a function of distance from the plant for two scenarios. One with only wet deposition and the other with only dry deposition.

This result indicates that over distances of several kilometers the relative concentration of RGM will change significantly as a result of deposition processes. Unfortunately the accuracy and positioning of the measurements performed during this period were not suitable to detect changes in relative concentrations.

4 Sensitivity simulations

The model calculations shown in the previous section are for a given set of emissions and deposition parameters. In order to test the sensitivity of the model results to changes in these parameters three sensitivity experiments have been carried out. These concentrate on GEM and RGM emissions and depositions, as these are the most significant contributors to deposition in the region close to the plant. These sensitivity tests are:

1. The sensitivity of the model to dry deposition velocities and meteorology
2. The sensitivity of the model to RGM wet deposition rates and precipitation
3. The sensitivity of the model to RGM emissions

4.1 Dry deposition rate sensitivity

The sensitivity of the model calculated depositions of GEM to changes in dry deposition velocity can be most simply calculated using a linear relationship between emissions and depositions. This can be done since deposition rates are low, 0.28% of the emissions, for the runs described in the Section 3.

As previously described the deposition rate is dependent on ground level concentration and deposition velocity. The ground level concentration is a function of dispersion and advection, which in turn is dependent upon meteorology. As a result the deposition rate of GEM varies according to meteorological conditions. High stability and low wind speeds will lead to higher ground level concentrations and thus deposition. The simplest method to estimate the variability of deposition is by looking at the hourly deposition rates and correlating these with wind speed.

This has been done, Figure 6, for the 5 day period where the % GEM deposition/emission has been plotted against the inverse of the wind speed. This is based on the total deposition within a radius of 5 km of the plant, as in the previous section. For wind speeds > 2 m/s there is a clear relationship but there is much scatter for lower wind speeds. This is primarily due to the influence of stability and the variability it can introduce.

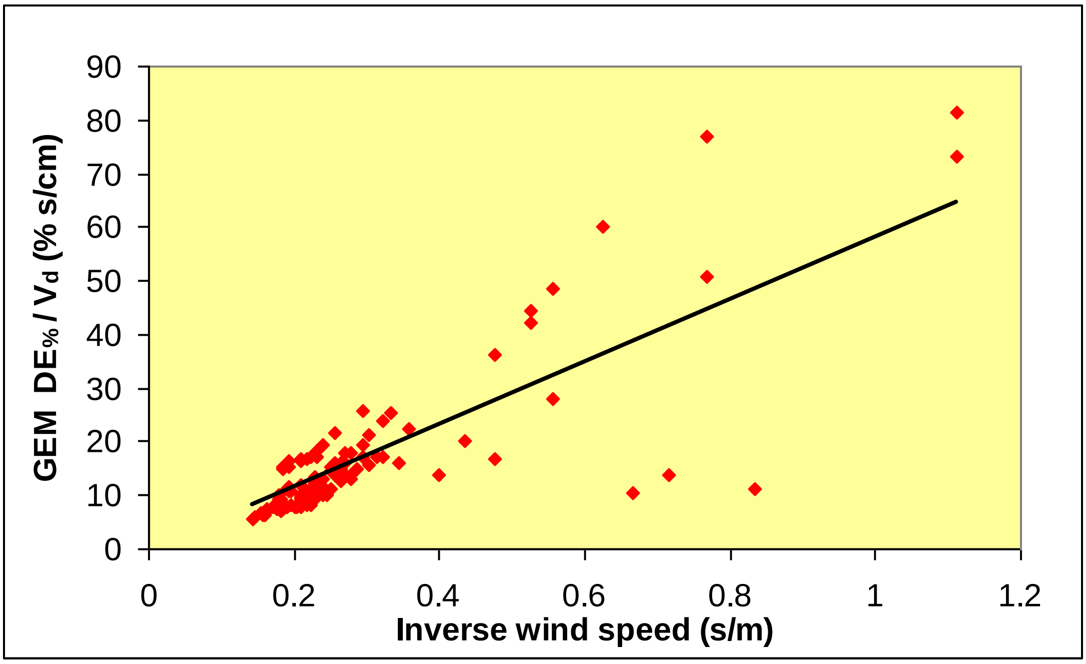


Figure 6. Plot showing the relationship between % GEM deposition/emission ($DE_{\%}$.) divided by dry deposition velocity (V_d) as a function of inverse wind speeds. The line of best fit, with a slope = 58, is also shown.

The results allow us to define the % ratio deposition/emission ($DE_{\%dry}$), as a function of dry deposition velocity (V_d) and wind speed (U) with the following equation.

$$DE_{\%dry} = 58\, V_d\, U^{-1}$$

where V_d is in units of cm/s and U is in units of m/s. This formulation is only expected to be valid for higher wind speeds, $U > 1$ m/s, and when deposition is not too large as to cause serious depletion of the plume, e.g. $DE_{\%dry} < 20$ %. It is also valid for RGM dry deposition when wet deposition does not occur.

This formulation is an approximation, since more information would be required concerning the dispersion characteristics, however it does indicate the basic sensitivity of deposition on wind speed and dry deposition velocity. It is not valid when emissions themselves are wind dependent. When this occurs, as with the plant in Rosignano, then we expect the dry deposition rates to be fairly constant. To test the robustness of such a description a longer simulation period would be needed. This will be done in the final EMECAP report where seasonal runs will be made.

4.2 Wet deposition rate sensitivity

The wet deposition rate is directly dependent on the precipitation, both its intensity and duration, as well as wind speed and the model defined wet scavenging ratio. To test the sensitivity of the simulated wet deposition rate to these parameters a number of scenario runs were carried out using differing wet scavenging ratios. The hourly results for 3 different simulations with differing wet scavenging ratios are shown in figure 7 as a function of time.

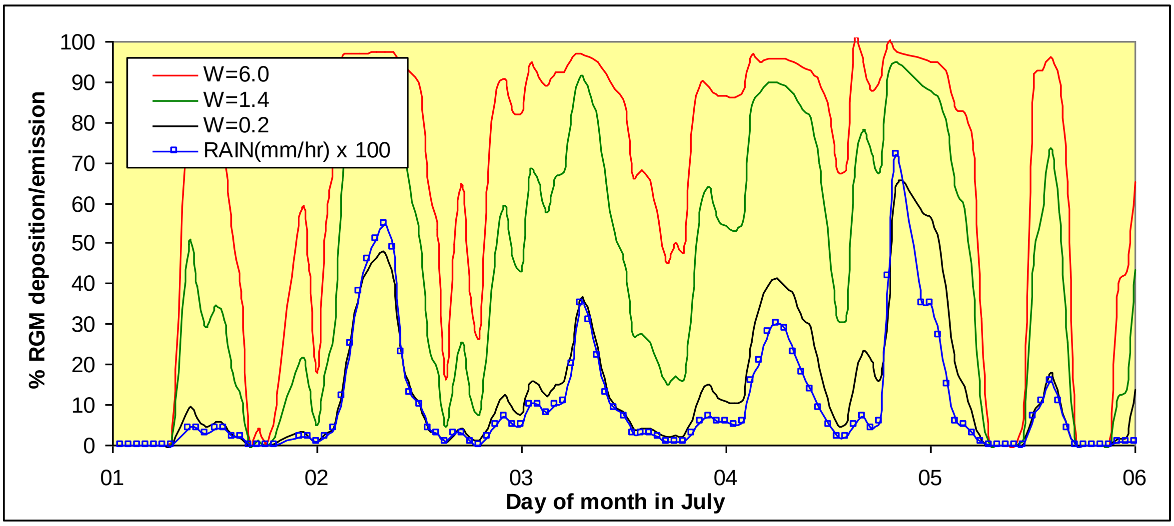


Figure 7 % of RGM emissions wet deposited within a 5 km radius of the plant during the 5 day scenario period. Shown are 3 of the scenarios where the wet scavenging ratio (W) is given as 6.0 $\times 10^6$, 1.4 $\times 10^6$ and 0.2 $\times 10^6$. Also shown is the TAPM generated precipitation (x 100) during the period in mm/hr.

The wet deposition follows chiefly the precipitation rate but is also influenced by wind speed, which can alter concentrations in the region. When rainfall is heavy then deposition can reach its maximum of 100%, independent of the wet scavenging ratio.

In figure 8 these data are plotted as a function of *P.W/U*, where *P* is precipitation (mm/hr), *W* is the wet scavenging ratio and *U* is the wind speed.

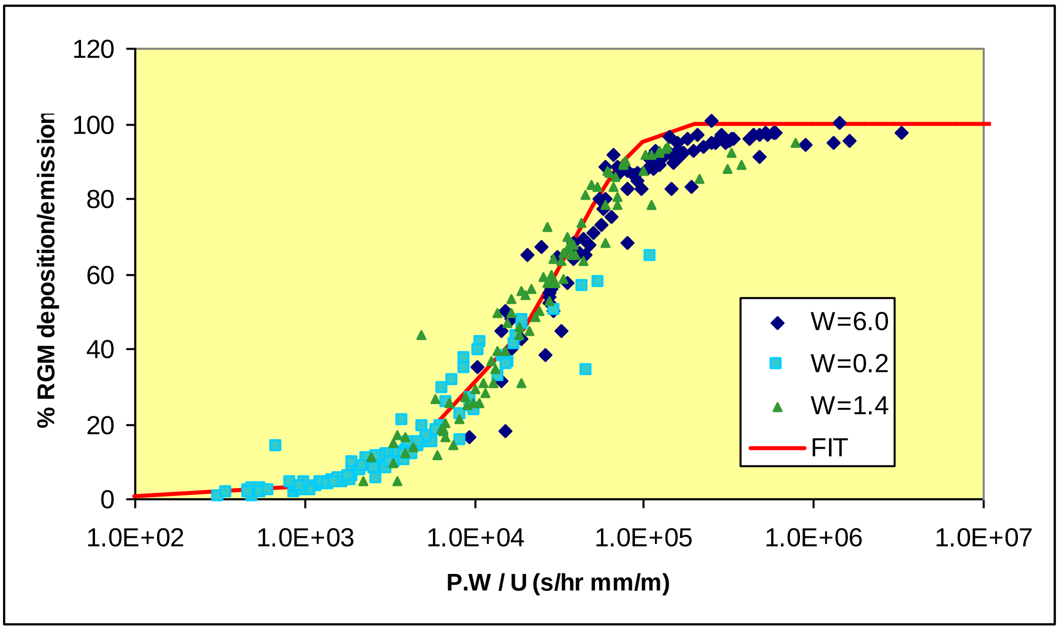


Figure 8 % of RGM emissions wet deposited within a 5 km radius of the plant during the 5 day scenario period. Shown are 3 of the scenarios where the wet scavenging ratio (W) is given as 6.0 $\times 10^6$, 1.4 $\times 10^6$ and 0.2 $\times 10^6$. This time the data are shown as a function of P.W/U. Also shown in red is the line of best fit.

The relationship, based on the wet deposition equation in Section 2.4, between the variable P.W/U will be an exponential one and the line of best fit is also shown in figure 8. This is given as

$$DE_{\%wet}(t) = 100\left(1 - \exp(-3\times10^{-5}\frac{W.P(t)}{U(t)})\right)$$

In order to calculate the mean deposition over a period of time the non-linearity of the above equation requires that it's integral be determined based on the hourly values of precipitation and wind speed. The result is shown in figure 9 below as a function of the wet scavenging ratio for model scenarios with two different dry deposition velocities.

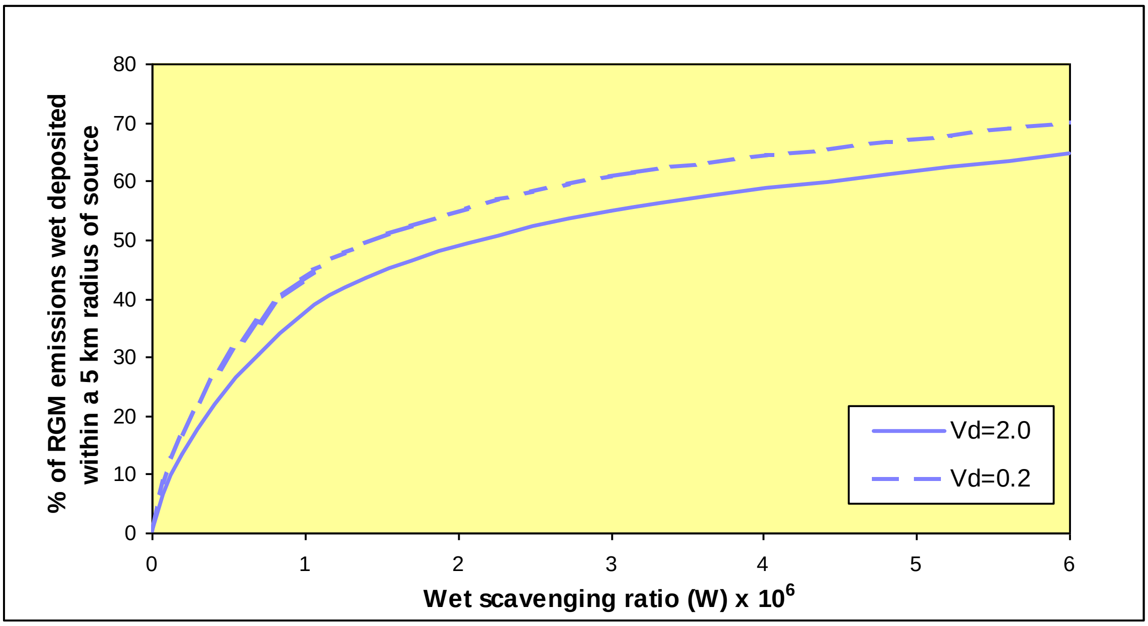


Figure 9. % of RGM emissions wet deposited within a 5 km radius of the plant during the 5 day scenario as a function of the wet scavenging ratio (W). Two different scenarios are shown with dry deposition velocities of 2.0 and 0.2 cm/s

As previously mentioned the period used for the sensitivity studies has an average rainfall rate similar to the yearly mean and so can be seen as representative in this regard. On the other hand rainfall duration was simulated to be 75% of the total period and this is not representative for the yearly duration.

The total wet deposition is reduced when the dry deposition velocity is higher as this depletes the plume. Figure 8 and 9 reflect the fact that increased scavenging exponentially approaches the maximum value of 100%, where all the compound will be washed out of the atmosphere. The maximum possible wet deposition will correspond to the d% duration of precipitation, which is 75% for this simulation.

From figure 9 it can be seen that the value of the wet scavenging ratio (1.4×10^6) used in this study is in an area of high sensitivity and uncertainty in this value can lead to significant variation in the total wet deposition.

4.3 Deposition sensitivity to RGM emissions

From the sensitivity runs carried out it is clear that the high rate of deposition of RGM can lead to a significant contribution of RGM deposition to the total Mercury deposition, even when RGM emissions are < 1% of the total Mercury emissions. As such it is important to see the effect that variations in RGM emissions have on the local deposition. In this sensitivity test the effect of varying RGM emissions from 0 to 50% of the total Mercury emissions is determined for the standard scenario run previously described. The results are displayed in figure 10.

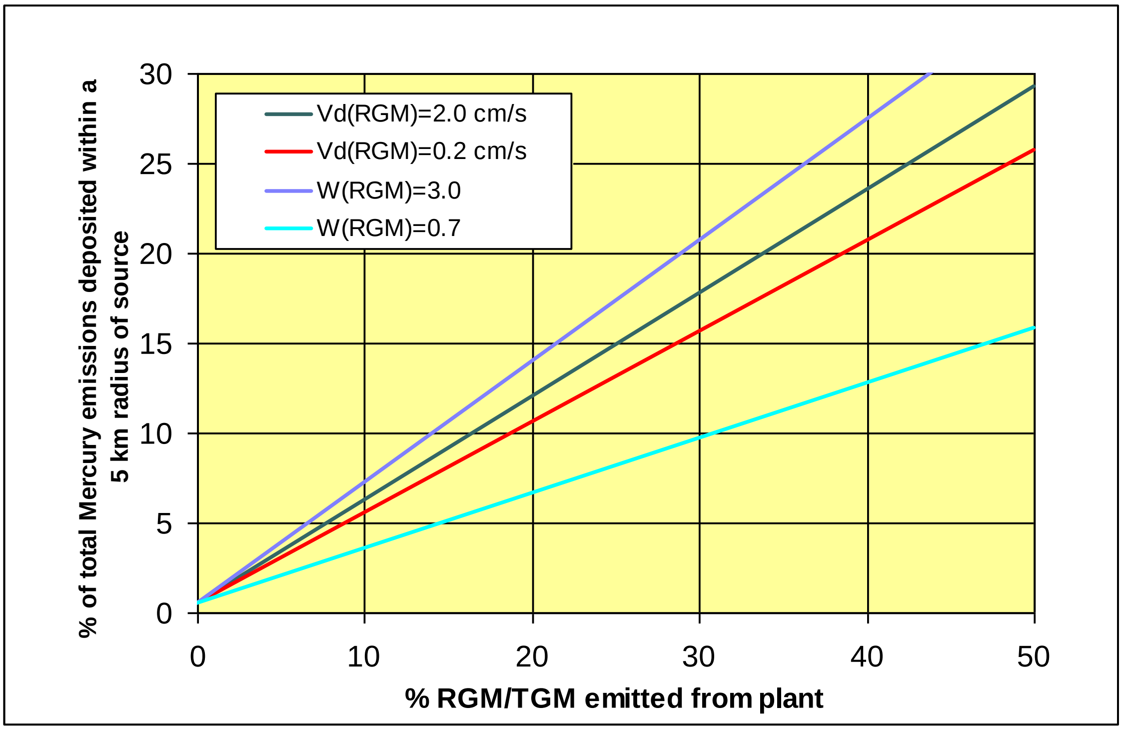


Figure 10 % of total mercury emissions deposited within a 5 km radius of the plant during the 5 day scenario as a function of the RGM/TGM emissions of the plant. 4 scenarios are shown with differing values of dry deposition velocities (Vd) and wet scavenging ratios ($W \times 10^6$).

In this figure 4 different scenarios are shown. Two with a constant wet scavenging ratio of 1.4×10^6 but with differing dry deposition velocities, 2.0 and 0.2 cm/s and two with a constant dry deposition velocity of 2.0 cm/s but with differing wet scavenging ratios, 3.0 and 0.7×10^6.

This figure shows the strong dependence of Mercury deposition on the RGM/TGM emission ratio for all 4 scenarios. As an example, using the standard scenario where V_d=2.0 cm/s and W=1.4×10^6, roughly 6% of all mercury emitted will be deposited in a 5 km radius when the RGM/TGM emission ratio is 10%. From table 5 this means that around 4% will be deposited within a 1 km radius. This is a factor of 15 higher than the expected deposition if all the Mercury were emitted in its elemental form.

Changes in the dry deposition velocity of the model do not significantly alter the deposition because of the significant rainfall during the simulation period and also due to the fact that when dry deposition is low wet deposition will increase, as more atmospheric Mercury will be available for scavenging.

It is evident that reduction of RGM emission can lead to a significant decrease in the local deposition of Mercury.

5 Conclusions and discussion

A number of scenario runs have been performed to test the sensitivity of the dispersion model to meteorology, deposition parameters and RGM emission ratios. Use has been made of a 5 day simulation period corresponding to the second EMECAP campaign period in Bohus. The results from these sensitivity studies have highlighted several points. These are:

1. The contribution of RGM to the local deposition, through both wet and dry deposition, is comparable to that of GEM even though RGM emissions make up < 1% of the total emissions at the Bohus plant.
2. Most of the Mercury, 60%, is deposited within a 1 km radius of the plant, and a large portion of this is through the deposition of RGM.
3. The total deposition of RGM is both wind speed and precipitation dependent.
4. Reductions in RGM emissions, when these are > 1% of the total emissions, can significantly reduce the local deposition of Mercury.
5. There are significant uncertainties in deposition velocities and wet scavenging rates for all Mercury species.

Though there is significant uncertainty in the deposition rates used, this is the current state of Mercury modelling. Improvements will only come through further well aimed experimetal campaigns that will help better define these deposition parameters.

It is worthy of note that reduction of RGM emissions will directly reduce the local deposition of Mercury. In the EMECAP report D5.2 it was suggested that the production of RGM is the result of gas phase reactions with Cl_2 within the plant. This reaction rate is dependent on Hg and Cl_2 concentrations as well as the dwell time in the plant. If this is the case, then the amount of RGM produced in the plant will be dependent on ventilation. RGM emissions may thus be signifcantly reduced simply by improving the ventilation within the plant.

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ABSTRACT

As part of Work Package 5 of the EU FP5 EMECAP project, dispersion model calculations have been carried out in order to determine the concentration and deposition of Mercury in the region surrounding 3 selected Mercury Cell Chlor-Alkali (MCCA) plants. In order to study the sensitivity of Mercury deposition in the local region surrounding the MCCA plants a number of sensitivity tests are carried out to determine local deposition around the plant as a function of dry deposition velocities, wet scavenging ratios and the percentage of Mercury emitted in the form of RGM. Sensitivity runs have been carried out at the Bohus plant in Sweden for a 5 day period. The results indicate that uncertainty in the deposition parameters, particularly the wet scavenging ratio, is significant in determining the total deposition. They also show that RGM is the most important species contributing to deposition despite its low representation, < 1% of TGM.

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