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# Statistical evaluation of the laboratory and field test data from the validation programme

Authors: Wenche Aas, Laurent Alleman, Elke Bieber, Dieter Gladtke, Vuokko Karlsson and Christian Monies

CEN/TC 264/WG 20 “Standard method for the determination of Pb, Cd, As and Ni in depositions“

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## Content:

# 1 Introduction

This report summarizes the results of the statistical evaluation of the lab- and field validation programme of CEN/TC264 WG20. This programme aims at the characterization of the reference method that is going to be described in the CEN standard "Standard methods for determination of lead, nickel, arsenic and cadmium atmospheric deposition".

The 4th Daughter Directive (EU, 2005) gives maximum allowable uncertainties in deposition measurements of arsenic, cadmium, nickel, mercury and polycyclic aromatic hydrocarbons (PAH). There are other CEN standards being developed for mercury (CEN/TC264/WG 25), PAH in air (prEN 15549) and for heavy metals in air (EN14902).

The 4th Daughter Directive (DD) states that the data quality objective for total deposition of Ni, Cd and As should have an uncertainty less than 70 %, expressed at a 95 % confidence level. Requirements for Pb measurements are given in the 1st Daughter directive (EU, 1999), but only for ambient air and not deposition. The data quality objectives for total deposition of Ni, Cd and As given in the 4th DD are therefore applied also for Pb. One should however notice that the Directive gives no limit values for deposition of heavy metals.

In addition, it is important to also consider the requirements given in the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) under the Convention on Long-range Transboundary Air Pollution (CLTRAP) where heavy metal deposition measurements are a mandatory part of the programme for the EMEP Parties. The EMEP data quality objectives (EMEP, 1996) states that the uncertainty in the rural background measurements of heavy metals should be less than 30% in the annual averages.

The 4th DD and EMEP have different requirements for methodology. The EU member states should use bulk collector for total deposition measurements, but wet only if difference between wet only and bulk is less than 10 %. The EMEP Parties should use wet only collector but bulk can be used if proven equivalent, meaning less than 10 % difference. It is important that the EU countries affected by both requirements are given proper guidelines on which collectors and methods are suitable to use in which environments to ensure that the heavy metal deposition measurements are valid for the different monitoring purposes.

The ISO/DIS 20988 (ISO, 2005) standard is used as a guideline to estimate the measurement uncertainty. The performance characteristics as repeatability and reproducibility have been determined and used for assessing the uncertainty in the measurements. This standard has implemented the general recommendations of the "guide to the expression of uncertainty in measurements" (GUM).

There are many sources of uncertainty in the deposition measurements and they depend on measurement methods, both in the laboratory and on field. The uncertainty also depends on the deposition load, whether the site is close to emission sources or mainly influence by long-range transport. The meteorology is of importance, e.g. whether it is

mainly wet- or dry conditions and if the deposition in winter comes with rain or snow. During the field trials, it has therefore been focused on covering all these different aspects on deposition monitoring.

The field validation takes into account all steps including sampling, preparation and analysis of the samples. Field validation tests were carried out at four measurement sites, one industrial, one urban and two rural; and five laboratories participated in the tests. The laboratories performance was tested using different type of intercomparison samples. For variation of the analytical method both graphite furnace AAS and ICP-MS were taken into account. To investigate the difference in deposition estimates, various types of collectors were uses, wet only (from Eigenbrodt), bulk (NILU type) and Bergerhoff type of gauge. The sampling campaign lasted for six months at each sampling site spanning different seasons between January 2006 and April 2007. Parallel measurements of two wet-only samplers, two bulk samplers and four Bergerhoff samplers were used. 20 % of the samples were split between laboratories for inter-laboratory reproducibility purpose.

## 2 Calculation of uncertainty

Following the recommendation from EN-ISO 20988, the standard uncertainty is estimated using parallel independent measurements and in order to calculate the between sampler uncertainty formula, following the experimental design and valuation method A8. This is a direct approach including variations and uncorrected biases together. The standard formula for paired measurements system is described in (1) and (2):

For wet only and bulk (two parallel sampler):

$$\mathrm{var}(y) = \sum_{j=1}^{N} \frac{(y_{(1,j)} - y_{(2,j)})^2}{2N} \qquad SD = \sqrt{\mathrm{var}(y)} \tag{1}$$

where

| | |
|---|---|
| var | = variance |
| SD | = standard deviation |
| $y_{(1,j)}$ | = observed value in parallel 1 |
| $y_{(2,j)}$ | = observed value in parallel 2 |
| N | = number of parallel samples |

For Bergerhoff (three parallel sampler):

$$\mathrm{var}(y) = \frac{\sum_{i=1}^{n}\sum_{j=1}^{p}(y_{i,j} - \bar{y}_i)^2}{N(P-1)} \qquad SD = \sqrt{\mathrm{var}(y)} \tag{2}$$

where

| | |
|---|---|
| var | = variance |
| SD | = standard deviation |

$y_{i,j}$ = observed value in period *i* in *j* measurement

$\bar{y}_i$ = average value for all the replicates/parallels

N = number of parallel samples

P = number of replicates/parallel samples

Equation (1) is also used for calculating the standard deviation between laboratories and when comparing the different type of samplers.

These calculations are used for evaluating the uncertainty in the measurements. To obtain the expanded uncertainty expressed at the 95 % confidence interval, the standard deviation is multiplied with a coverage factor (k) of 2:

Expanded uncertainty = k* SD, where k= 2.

# 3 Laboratory validation test

The laboratory test were carried out before the field test trials in order to show that the participating laboratories were able to perform the analysis of heavy metals with the required precision, and to obtain information about the detection limits of the methods.

5 labs participated in the laboratory trials.

LUA: Landesumweltamt Nordrhein-Westfalen, Essen, Germany.
NILU: Norwegian Institute for Air Research, Kjeller, Norway
UBA: Umweltbundesamt, Langen, Germany
FMI: Finnish Meteorological Institute, Helsinki, Finland
EMD: Ecole des Mines de Douai, Douai, France

Analyses were carried out by ICP-MS (EMD, NILU, FMI, and UBA) or graphite furnace AAS (LUA) according to EN 14902. Digestions of solid samples were carried out using the microwave technique according to the same EN 14902.

4 different samples were analysed.

**Rain water blank**: This was prepared by each laboratory from acidified deionised water
**Solid blank**: concentrated $HNO_3/H_2O_2$ evaporated and diluted with deionised water as used for the digestion of Bergerhoff samples.
**Synthetic heavy metal samples:** from EMEP laboratory intercomparison program 2004
**SRM and NIES** for the digestion and analysis of the metals
**Solid deposition sample** from Bottrop, already evaporated

The detection limit is defined as the threefold standard deviation of the mean blank samples. The detection limit for the precipitation samples are given in **Table 1**.

**Table 1.** Detection limits for heavy metals (3x SD of blank), in μg/L.

| Rain water blank | NILU | FMI | UBA | LUA | EMD |
|---|---|---|---|---|---|
| As | 0.0030 | 0.0079 | 0.0024 | 1.20 | 0.003 |
| Cd | 0.0003 | 0.0012 | 0.0018 | 0.01 | 0.001 |
| Ni | 0.0033 | <0.02 | 0.0040 | 0.09 | 0.003 |
| Pb | 0.0007 | <0.03 | 0.0012 | 0.20 | 0.001 |

These measurements are not absolute values, but representative for the intercomparison. The detection limit does vary but it is not expected to change substantially from the lab test to the field campaign period.

The detection limits at the LUA lab are significantly higher than those of the other laboratories, due to the relatively lower precision of graphite furnace AAS used by LUA compared to ICP-MS, the analytical technique used by the other laboratories.

To evaluate the detection limit on deposition in $\mu g/m^2$ it is necessary to estimate the lowest precipitation amount possible to sample and detect. This is somewhat dependent on the sample collector. The larger the opening surface, the more precipitation is possibly collected and therefore, less rain amount in mm is needed. Therefore only detection limit in μg/L is given above.

For the solid samples, evaporation as well as a dilution step is included in the preparation of these blanks, the values are therefore higher than those of the rainwater samples, as shown in **Table 2**. During the field trial, only LUA and EMD measure solid samples, but in addition, UBA and FMI have participated in the laboratory validation intercomparison

**Table 2.** Detection limits of solid samples (3x SD of blank), μg sample.

| solid sample | FMI | UBA | LUA | EMD |
|---|---|---|---|---|
| As | 0.0020 | 0.0008 | 0.71 | 0.039 |
| Cd | 0.0002 | 0.0001 | 0.03 | 0.024 |
| Ni | 1.10 | 0.01 | 0.57 | 1.53 |
| Pb | 0.003 | 0.003 | 0.49 | 1.09 |

The EMEP laboratory intercomparison consists of 4 samples, where two samples are representing precipitation in low polluted areas in Europe. Two samples with higher levels can represent more urbanized or industrial areas. Results of artificial rainwater samples are summarized in **Table 3**. In this intercomparison a sample used for the annual laboratory intercomparison in EMEP was distributed to the WG20 laboratories. Three of these labs also participate in the regular EMEP intercomparison that consists of the same samples. Results from both these analyses are presented in the table.

Due to the low precision of graphite furnace AAS the results of LUA for the low contaminated samples differ significantly from those of the other laboratories using the ICP-MS technique. The results from the low concentrations for LUA are eliminated from the calculation of the standard deviation. For high concentration samples, the GF AAS works fine. The standard deviation between the different laboratories is below 5%

showing that all the labs perform well. When compared with the expected value, which is the theoretical value calculated from the amount of metals used for preparing the samples, the results are also quite good. For As and Cd, all the labs present deviations below 6 %. There are somewhat higher errors in the Ni measurements and this can be due to contamination. It is relatively easy to contaminate the sample with Ni. For Pb, NILU and partly FMI had relatively high error in the high concentration samples, when looking at how these two labs performed in the regular EMEP intercomparison it was much better, see **Table 3**.

**Table 3.** Results of the analysis of artificial rain water samples, μg/L.

| | As | | | | Cd | | | | Pb | | | | Ni | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample id | H1 | H2 | H3 | H4 | H1 | H2 | H3 | H4 | H1 | H2 | H3 | H4 | H1 | H2 | H3 | H4 |
| NILU | 0.69 | 0.78 | 5.06 | 6.03 | 0.060 | 0.082 | 0.902 | 0.692 | 1.31 | 1.60 | 43.1 | 17.2 | 0.67 | 0.75 | 6.32 | 7.28 |
| FMI | 0.67 | 0.78 | 5.03 | 5.76 | 0.056 | 0.076 | 0.880 | 0.658 | 1.19 | 1.51 | 46.7 | 18.2 | 0.65 | 0.75 | 7.04 | 7.69 |
| UBA | 0.71 | 0.84 | 5.23 | 6.27 | 0.058 | 0.078 | 0.905 | 0.688 | 1.30 | 1.55 | 48.2 | 19.2 | 0.70 | 0.79 | 7.02 | 7.67 |
| EMD | 0.67 | 0.77 | 4.67 | 5.75 | 0.060 | 0.079 | 0.873 | 0.682 | 1.28 | 1.55 | 47.1 | 18.8 | 0.72 | 0.83 | 6.84 | 7.74 |
| LUA | <DL | <DL | 5.29 | 6.35 | <DL | 0.07 | 0.855 | 0.66 | <DL | <DL | 49.3 | 17.5 | <DL | <DL | 6.74 | 7.75 |
| SD% | 3.0 | 4.1 | 4.8 | 4.7 | 3.3 | 3.2 | 2.3 | 2.3 | 4.4 | 2.5 | 5.1 | 4.6 | 4.5 | 4.9 | 4.3 | 2.6 |
| | | | | | | | | | | | | | | | | |
| **EMEP intercomparison** | | | | | | | | | | | | | | | | |
| NILU | 0.70 | 0.80 | 4.88 | 5.93 | 0.062 | 0.083 | 0.907 | 0.715 | 1.32 | 1.63 | 48.6 | 20.33 | 0.75 | 0.82 | 6.31 | 7.38 |
| FMI | 0.68 | 0.79 | 4.92 | 5.81 | 0.059 | 0.081 | 0.888 | 0.678 | 1.22 | 1.51 | 48.8 | 20.10 | 0.59 | 0.69 | 6.50 | 7.30 |
| UBA | 0.71 | 0.82 | 5.08 | 6.12 | 0.057 | 0.077 | 0.879 | 0.686 | 1.26 | 1.54 | 47.2 | 19.20 | 0.68 | 0.75 | 6.60 | 7.59 |
| Exp.value | 0.70 | 0.80 | 5.00 | 6.00 | 0.06 | 0.08 | 0.9 | 0.7 | 1.30 | 1.60 | 50.0 | 20.00 | 0.70 | 0.80 | 7.00 | 8.00 |

**Average percent error (absolute values) from expected value:**

| | **As** | | **Cd** | | **Pb** | | **Ni** | |
|---|---|---|---|---|---|---|---|---|
| | low | high | low | high | low | high | low | high |
| NILU | 2 % | 1 % | 1 % | 0.2% | 1 % | 14 % | 5 % | 9 % |
| FMI | 3 % | 2 % | 6 % | 2 % | 7 % | 8 % | 7 % | 2 % |
| UBA | 3 % | 4 % | 3 % | 1 % | 2 % | 4 % | 1 % | 2 % |
| EMD | 4 % | 5 % | 1 % | 3 % | 2 % | 6 % | 3 % | 3 % |
| LUA | | 6 % | | 5 % | 6 % | 7 % | | 3 % |
| **EMEP intercomparison:** | | | | | | | | |
| NILU | 0.3 % | 2 % | 4 % | 1 % | 2 % | 2 % | 5 % | 9 % |
| FMI | 2 % | 2 % | 1 % | 1 % | 6 % | 1 % | 15 % | 8 % |
| UBA | 2 % | 2 % | 4 % | 2 % | 3 % | 5 % | 5 % | 5 % |

Solid samples from Bottrop were distributed to four labs, LUA, EMD, FMI and UBA. The two latter did not take part in the field trial but took part as volunteers in the lab intercomparison. The participating laboratories digested and analysed the material according to EN 14902. The solid deposition samples results are summarised in **Table 4**.

Compared to the rainwater samples the inter laboratory differences are far higher. The reason for this is that more work up steps are included in the preparation of the samples: homogenisation of a slurry containing solid material, evaporation and acid digestion, each of these steps leading to a higher uncertainty of the result. An additional uncertainty in this exercise is that not exactly the same powder is sent to each lab

because different sampler are used to sample all the filter material necessary for the analysis. The quantity of material used in each lab is also an important factor as the weighting uncertainty as well as heterogeneity of the solid sample adds to the uncertainty. The standard deviations of one single laboratory are by far lower as expected since this is just repeatability. These data are calculated from one digested sample analyzed five times.

**Table 4.** Results of the analysis of solid deposition samples from Bottrop, µg.

| | FMI | EMD | UBA | LUA | SD |
|---|---|---|---|---|---|
| mean As | 0.66 | 0.63 | 0.72 | 0.52 | 12 % |
| SD | 5 % | 2 % | 1 % | 6 % | |
| Mean Cd | 0.12 | 0.13 | 0.13 | 0.10 | 8 % |
| SD | 5 % | 2 % | 4 % | 1 % | |
| mean Ni | 3.70 | 5.21 | 6.12 | 4.82 | 22 % |
| Sdi | 3 % | 1 % | 1 % | 2 % | |
| mean Pb | 5.53 | 6.83 | 7.41 | 4.84 | 19 % |
| SD | 3 % | 1 % | 1 % | 1 % | |

# 4 Sites description for the field validation trials

Four different sites were selected, 1 industrial, 1 urban, 1 remote southern and 1 remote northern site. The laboratories presented in the introduction were analysing various samples defined in the set-up given below for each site. Parallel sampling of the different collectors were analysed. It was two wet-only samplers, two bulk samplers and four Bergerhoff samplers at each site. Each measurement site was responsible for operating all samplers and also for delivering the samples to the other participating labs. The sampling campaigns were lasting at least six months at each sampling site. The sampling periods for wet-only and bulk collectors are one week and for Bergerhoff collector four weeks.

Additionally, quality control samples such as field and laboratory blanks were analysed. At least 20 % of the parallel samples were shipped to one other laboratory for analysis. That means that 1 of 4 Bergerhoff samplers was analysed in one lab while 3 in another. For the parallel bulk and wet only sampler, one sampler was sent to another lab every month. In addition, some additional split samples were analysed at two different laboratories. Details of the measurement programme and methods are described in the guideline document for the field trials (CEN/TC264/WG20 N025).

The 4 different sites have very different deposition loads of heavy metals. The highest levels were expected at the industrial site in Duisburg, Germany and the lowest at the remote sites in Norway (Birkenes) and France (Peyrusse). The urban site in Copenhagen was expected to have somewhat higher level than the rural sites, but deposition measurements have not been performed here previously. From earlier measurements it is seen that the deposition level is about one order of magnitude higher in Duisburg compared to Birkenes, see **Table 5**. At Duisburg, the deposition is estimated as average of the four closest deposition measuring sites using Bergerhoff collector. At Birkenes the results are wet deposition using bulk collector. The total deposition at this site consists however of mainly wet deposition, between 70 % - 90 %.

**Table 5.** Deposition of heavy metals in Birkenes (bulk deposition) and Duisburg (Bergerhoff deposition) in 2004 µg/(m²d).

| | Birkenes | Duisburg |
|---|---|---|
| Cd | 0.2 | 1 |
| Pb | 6 | 50 |
| As | 0.6 | 2.3 |
| Ni | 1 | 20 – 50 |
| mm | 1709 | 910 |

More details of the different sites and measurement programmes are described in the next chapters.

### 4.1 Industrial site: Duisburg Meiderich, Germany

The Duisburg site (EU code: DENW037) is located in a residential area with a high population density. Duisburg harbour (51° 31´N, 6° 58´E, 30 masl), the largest inland harbour of Europe is situated 500 m southwest of the site. As it is shown in **Figure 1** the site is surrounded by smaller metal processing factories, huge iron and steel producing factories are situated in western to north-westerly direction in a distance of 1 km to 2 km. Most abundant wind direction: south-west.

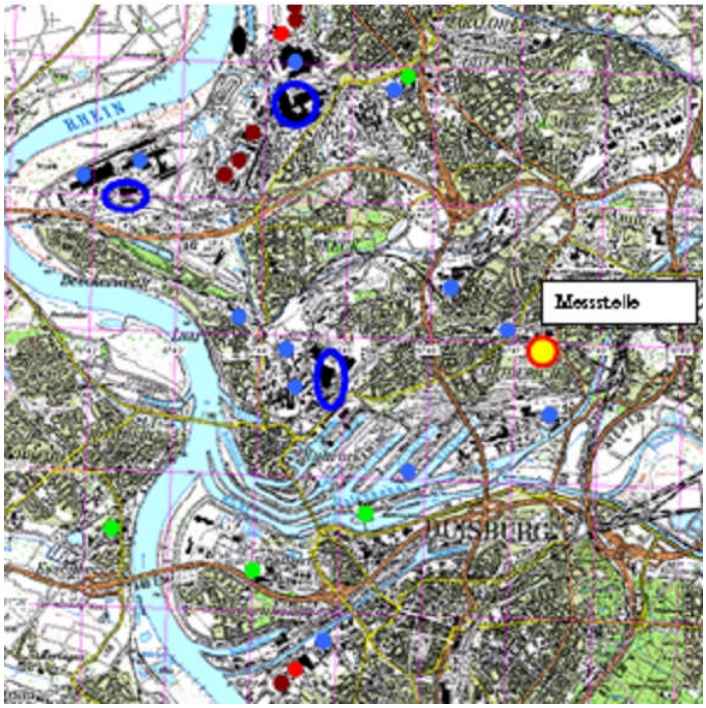


**Figure 1**. Shows a map with the location of the station and industry nearby.

Blue dots metal processing factories
Green dots chemical industry
Brown dots blast furnaces

Red dots sintering plants
Blue ellipses steel mills
Black dot cookery

The field validation was carried out between 16th of January and 4th of July 2006. The devices were placed within a shielded area as illustrated in **Figure 2**.

**Figure 2.** Deposition measurements at Duisburg.

Four cylindrical gauges (Bergerhoff), two wet only samplers (Eigenbrodt) and two bulk (NILU type) samplers were installed at the site. The bottles of the wet only and the bulk samplers were changed and analysed weekly, their funnels were also analysed weekly at the first month, after that they were analysed monthly. The cylindrical gauges were changed and analysed on a monthly basis.

At the site the responsible laboratories for the various collectors were as follows:

FMI Bulk collector (NILU type), 20 % of the samples were split an analysed at UBA
LUA 3 Bergerhoff cylindrical gauge
EMD 1 Bergerhoff cylindrical gauge
UBA Wet only (Eigenbrodt), 20 % of samples were split and analysed at FMI

### 4.2 Urban site: Copenhagen, Denmark

The site in Copenhagen (55°41'N, 12°34'E) is in the botanic garden, see **Figure 3**, and the influence of nearby sources is minimal. The site can be characterized as urban background. The field intercomparison was carried out between 5th of September 2006 and 20th of February 2007.

**Figure 3.** Deposition measurements in Copenhagen.

At the site the responsible laboratories for the various collectors were as follows:

FMI Bulk collector (NILU type), 20% of samples were analysed at UBA
LUA 1 Bergerhoff cylindrical gauge
EMD 3 Bergerhoff cylindrical gauge
UBA Wet only (Eigenbrodt), 20% of samples were analysed at FMI

### 4.3 Southern remote site: Peyrusse, France

The site (43º 37'N, 00º 11'E, 236 masl) is near the village of Peyrusse Vieille in a region where valley networks are separated by forested hills with slow inclination, see **Figure 4**. Easterly winds normally prevail. The site is far away from urban or industrial sources of pollution and it is part of the EMEP network. The field intercomparison was carried out between 7$^{th}$ of March to 22$^{nd}$ of August 2006.

**Figure 4.** Deposition measurements in Peyrusse.

At the site the responsible laboratories for the various collectors were as follows:
NILU Bulk collector (NILU type), 20 % of samples were split and analysed at UBA
LUA 1 Bergerhoff cylindrical gauge
EMD 3 Bergerhoff cylindrical gauge
UBA Wet only (Eigenbrodt), 20 % of samples were split and analysed at NILU

### 4.4 Northern remote site: Birkenes, Norway

The site (58º 23'N, 8º 15'E. 190 masl) is a regional site with minimal local sources. The heavy metal exposure is mainly from the European continent. The site is situated in a forested area, the terrain is undulating and the site is located in a clearing with relatively free exposure to exchange of air masses by wind, see **Figure 5**.

**Figure 5.** Deposition measurements at Birkenes.

At the site the responsible laboratories for the various collectors were as follows:

NILU Bulk collector, 20 % of samples were analysed at UBA
LUA 1 Bergerhoff cylindrical gauge
EMD 3 Bergerhoff cylindrical gauge
UBA Wet only (Eigenbrodt), 20 % of samples were analysed at NILU

## 5 Results from field trials

### 5.1 Introduction

There are different approaches to estimate deposition. Wet deposition is defined as the amount being scavenged by precipitation. The precipitation found in the wet only collector is the best approach to get the wet deposition. However some of the heavy metals in the precipitation might get absorbed on the funnel walls, and to estimate the importance of this effect, the funnel was rinsed with acid every month and the rinsing water was analysed for heavy metals. When using a bulk collector, one will get wet deposition plus some contribution of dry deposition depending on the meteorology and particle concentration at the site. To get an estimate of the contribution of dry deposition to the bulk deposition on the funnels, they are rinsed with acidic water every week

(Duisburg) or every fourth week (the three other sites), and the rinsing water is analysed. As for the wet only collector, some of the heavy metals in the bulk precipitation are absorbed on the funnel walls in addition there is contribution from dry deposition. For the Bergerhoff collectors, the sampler collects both wet and some dry deposition. However one should notice that neither the bulk+funnel nor the Bergerhoff sampler collects total deposition. The precipitation collectors do not capture the total dry depositions from small particles.

The deposition in each wet only and bulk collector was calculates as the product of precipitation amount in mm and the concentration in µg/L. The precipitation amount was calculated from the amount in ml and adjusted for the size of the funnel. The deposition on the funnel wall is calculated similar using the amount of ml rinsing solution. The deposition was then averaged for the number of days the measurement period lasted. Data below the detection limit are given as half the detection limit. At Birkenes there was problems with overflow of the wet only and Bergerhoff collectors. For the wet only the precipitation amount from the bulk collector was used in the weeks where this was a problem. It is not corrected for outliers and there is no correction for field blanks.

In **Figure 6** an overview of the deposition in the different collectors, including what observed from the funnel wash, from all the field trial sites are shown. The parallel samples are averaged to get one measure for each collector type. For the Bergerhoff sampler only the three parallel samples analysed by the same laboratory is used.

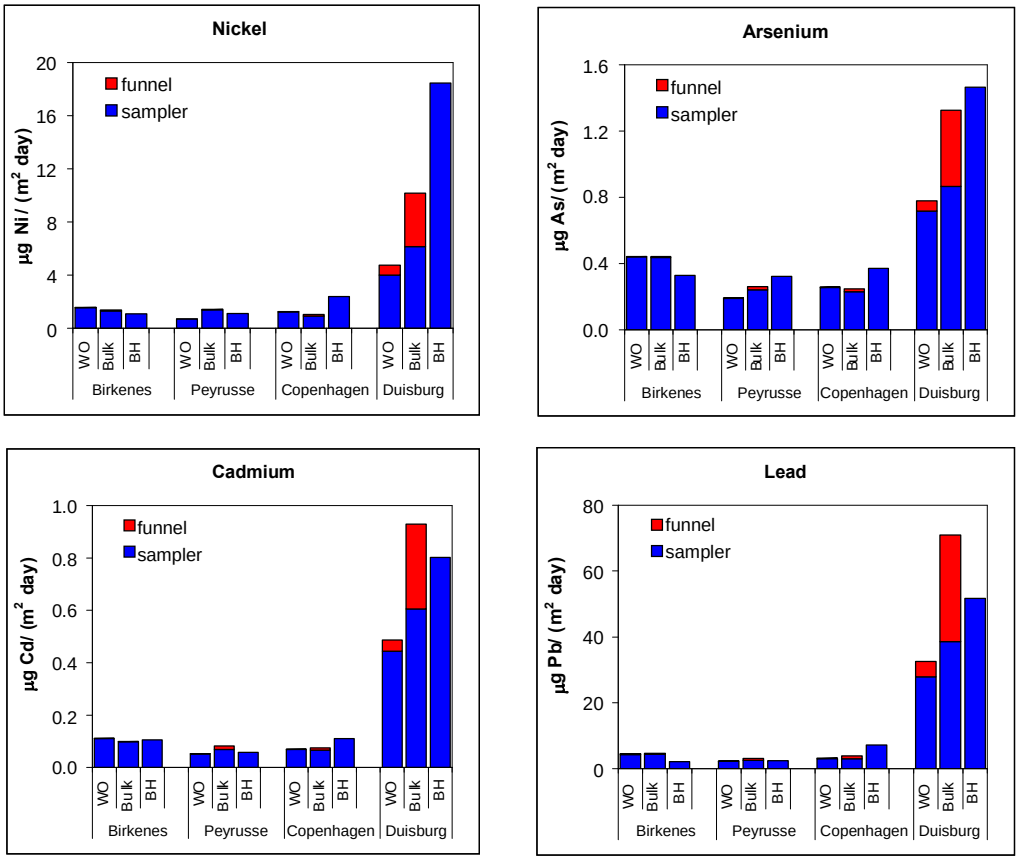


**Figure 6.** Deposition of As, Cd, Ni and Pb at the four field sites using three different collectors, WO means wet only and BH Bergerhoff.

It is very clear from **Figure 6** that the deposition at the industrial site in Duisburg is much higher than the three other sites, which all are on the same level. The results from Copenhagen show similar values as the two remote sites, indication that Copenhagen is not very polluted with heavy metals. This might be the case for many European cities. It is only at Duisburg the funnel wash contributes significantly, both to the wet only and bulk samplers. Peyrusse has somewhat higher contribution to the funnel in the bulk collector than at the other sites; this can be caused by a drier climate at Peyrusse than at the Scandinavian sites, which experience more frequent rain events. There are differences between sites also when comparing the wet only and bulk collectors. The wet deposition at Birkenes and Copenhagen are very similar for these two collectors indicating that at sites with frequent rain events mainly experience wet deposition of heavy metals. At Peyrusse and Duisburg the deposition in the bulk collector is enhanced, especially true for Duisburg, indicating that dry deposition has contributed significantly. At polluted and/or dry sites it is therefore recommended to use a wet only to estimate the wet deposition. The Bergerhoff collector show sometimes higher and sometimes lower deposition loads than the two other collectors, which might be due to somewhat higher uncertainty in these measurements and some problems with overflow of the collector, see the below sections for details. But generally it seems like Bergerhoff and bulk+funnel deposition calculations are reproducing similar results.

Since all the measurements have been run in parallel there are several types of uncertainties that can be calculated. The between sampler uncertainty are calculated between the same type of collector as well as between collectors that, in principle, should give similar results, i.e. wet only and bulk; and Bergerhoff and bulk+funnel. In addition, several samples have been split between the laboratories to estimate the inter-laboratory uncertainty. Within this estimate, uncertainty in transport is also included. At the Copenhagen and Birkenes intercomparison 20% of the parallel wet only and bulk samples were sent directly to the second laboratory before digestion or other sample preparation procedures. The intercomparison at these sites also includes the uncertainty in sample preparation. Measurements below the detection limits were not used in the uncertainty estimates. In the calculations only periods with parallel sampling are used in the deposition and uncertainty estimates, there might therefore be some minor difference between the numbers given in the tables below and what shown in **Figure 6**. When there were problems with one of the precipitation collector, i.e overflow or other problems the deposition was calculated using the precipitation amount from the other parallel sampler. But otherwise the precipitation amount is part of the total uncertainty in the estimates. All the raw data used in the statistical evaluations are found in the Appendix.

### 5.2 Between sampler uncertainty

The bulk collectors were operating for about 20 weeks at all the sites and the funnel wash were done every $4^{th}$ week. For the collector + funnel there are therefore only up to six parallel sample periods to compare, which gives a rather uncertain estimate of the standard deviation. At Peyrusse, the funnel wash was only done during the last three months so the data on deposition average from the bulk only and the bulk + funnel is not directly comparable at this site. For the Bergerhoff intercomparison the between sampler uncertainty is based on three parallel samples, the fourth sample is used for the

between laboratory uncertainty discussed in the next chapter. A summary of the statistical calculations is found in **Table 6** to **Table 8**.

**Table 6.** Average deposition and the relative standard deviation in the bulk measurements.

**Birkenes**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.10 | ± | 0.01 | 11 % | 22 |
| Pb | 4.2 | ± | 0.6 | 13 % | 22 |
| As | 0.42 | ± | 0.09 | 22 % | 21 |
| Ni | 1.3 | ± | 0.6 | 48 % | 19 |
| mm | 34.2 | ± | 2.9 | 9 % | 20 |

| bulk + funnel dep, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.096 | ± | 0.006 | 6 % | 6 |
| Pb | 4.4 | ± | 0.4 | 9 % | 6 |
| As | 0.42 | ± | 0.04 | 9 % | 6 |
| Ni | 1.4 | ± | 0.4 | 30 % | 6 |

**Peyrusse**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.07 | ± | 0.02 | 32 % | 16 |
| Pb | 2.6 | ± | 0.5 | 20 % | 16 |
| As | 0.25 | ± | 0.04 | 17 % | 16 |
| Ni | 1.5 | ± | 0.2 | 16 % | 16 |
| mm | 16.5 | ± | 1.6 | 10 % | 16 |

| bulk + funnel dep, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.08 | ± | 0.02 | 22 % | 3 |
| Pb | 2.9 | ± | 0.2 | 8 % | 3 |
| As | 0.23 | ± | 0.02 | 10 % | 3 |
| Ni | 1.3 | ± | 0.3 | 21 % | 3 |

**Copenhagen**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.07 | ± | 0.01 | 19 % | 20 |
| Pb | 3.0 | ± | 0.4 | 13 % | 20 |
| As | 0.22 | ± | 0.02 | 9 % | 20 |
| Ni | 0.9 | ± | 0.1 | 15 % | 20 |
| mm | 14.8 | ± | 0.2 | 2 % | 22 |

| bulk + funnel dep, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.068 | ± | 0.007 | 11 % | 6 |
| Pb | 3.9 | ± | 0.2 | 6 % | 6 |
| As | 0.24 | ± | 0.01 | 5 % | 6 |
| Ni | 1.0 | ± | 0.0 | 5 % | 6 |

**Duisburg**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.57 | ± | 0.07 | 13 % | 21 |
| Pb | 36.3 | ± | 4.8 | 13 % | 21 |
| As | 0.82 | ± | 0.07 | 9 % | 21 |
| Ni | 5.8 | ± | 0.7 | 12 % | 21 |
| mm | 16.1 | ± | 1.3 | 8 % | 21 |

| bulk + funnel dep, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.90 | ± | 0.03 | 3 % | 6 |
| Pb | 68.8 | ± | 3.2 | 5 % | 6 |
| As | 1.30 | ± | 0.05 | 4 % | 6 |
| Ni | 9.8 | ± | 0.5 | 5 % | 6 |

As was seen in **Figure 6** the difference in bulk deposition, including funnel wash or not is minor except at the site in Duisburg, see **Table 6**. The relative uncertainty is lower in the bulk + funnel estimate, but these are calculated from very few samples and the uncertainty estimate is more robust for the bulk deposition only. There is no clear difference between the sites, except that the uncertainty is somewhat lower at the Duisburg site. This is probably due to the higher concentration level, which is easier to measure as well as that small contamination events has little impact.

At the Birkenes site, one of the wet only collectors were out of function after three months, so parallel measurements for this sampler is only valid for half the period. At this site, there were also problems with overflow of the wet only collector(s) for several months. For these weeks the average precipitation amount from the bulk collectors were used for calculation deposition. The measurements done with the wet only collectors shows similar pattern as for the bulk collector, see **Table 7**.

**Table 7.** Average deposition and the relative standard deviation in the wet only measurement.

**Birkenes**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.18 | ± | 0.03 | 18 % | 11 |
| Pb | 7.4 | ± | 0.5 | 7 % | 11 |
| As | 0.7 | ± | 0.1 | 19 % | 11 |
| Ni | 2.8 | ± | 0.3 | 12 % | 10 |
| mm | 34.7 | ± | 3.1 | 9 % | 7 |

| wet only + funnel, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.18 | ± | 0.01 | 6 % | 3 |
| Pb | 7.7 | ± | 0.2 | 2 % | 3 |
| As | 0.75 | ± | 0.07 | 9 % | 3 |
| Ni | 2.8 | ± | 0.1 | 5 % | 3 |

**Peyrusse**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.05 | ± | 0.01 | 13 % | 19 |
| Pb | 2.2 | ± | 0.3 | 15 % | 19 |
| As | 0.18 | ± | 0.02 | 11 % | 19 |
| Ni | 0.8 | ± | 0.1 | 13 % | 19 |
| mm | 15.7 | ± | 1.6 | 10 % | 18 |

| wet only + funnel, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.08 | ± | 0.003 | 4 % | 6 |
| Pb | 3.4 | ± | 0.2 | 6 % | 6 |
| As | 0.28 | ± | 0.01 | 4 % | 6 |
| Ni | 1.02 | ± | 0.04 | 4 % | 6 |

**Copenhagen**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.07 | ± | 0.01 | 9 % | 17 |
| Pb | 3.1 | ± | 0.3 | 11 % | 17 |
| As | 0.26 | ± | 0.02 | 6 % | 17 |
| Ni | 1.3 | ± | 0.3 | 20 % | 14 |
| mm | 15.7 | ± | 0.5 | 3 % | 19 |

| wet only + funnel, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.08 | ± | 0.02 | 28 % | 6 |
| Pb | 3.4 | ± | 0.9 | 26 % | 6 |
| As | 0.3 | ± | 0.1 | 29 % | 6 |
| Ni | 1.3 | ± | 0.2 | 17 % | 6 |

**Duisburg**

| deposition, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.44 | ± | 0.04 | 9 % | 19 |
| Pb | 28 | ± | 3 | 11 % | 19 |
| As | 0.72 | ± | 0.05 | 7 % | 19 |
| Ni | 4.0 | ± | 0.4 | 11 % | 19 |
| mm | 16.4 | ± | 0.5 | 3 % | 19 |

| wet only + funnel, µg/m²/day | Avg | | SD | rel SD | nr |
|---|---|---|---|---|---|
| Cd | 0.50 | ± | 0.02 | 5 % | 6 |
| Pb | 33 | ± | 3 | 8 % | 6 |
| As | 0.83 | ± | 0.04 | 5 % | 6 |
| Ni | 4.8 | ± | 0.8 | 18 % | 6 |

The Bergerhoff samplers were 4 in parallel and measured every month. 3 of the samples were analysed at the same laboratory and are used in the calculation of between sampler uncertainties, see **Table 8**.

**Table 8.** Average deposition and the relative standard deviation in the Bergerhoff measurements.

**Birkenes**

| deposition, µg/m²/day | | | | | |
|---|---|---|---|---|---|
| | Avg | | SD | rel SD | nr |
| Cd | 0.10 | ± | 0.02 | 20 % | 6 |
| Pb | 2.1 | ± | 0.9 | 44 % | 6 |
| As | 0.3 | ± | 0.1 | 31 % | 6 |
| Ni | 1.1 | ± | 0.3 | 24 % | 6 |

**Peyrusse**

| deposition, µg/m²/day | | | | | |
|---|---|---|---|---|---|
| | Avg | | SD | rel SD | nr |
| Cd | 0.05 | ± | 0.01 | 12 % | 6 |
| Pb | 2.3 | ± | 0.2 | 11 % | 6 |
| As | 0.30 | ± | 0.03 | 9 % | 6 |
| Ni | 1.03 | ± | 0.09 | 9 % | 6 |

**Copenhagen**

| deposition, µg/m²/day | | | | | |
|---|---|---|---|---|---|
| | Avg | | SD | rel SD | nr |
| Cd | 0.11 | ± | 0.03 | 28 % | 6 |
| Pb | 7 | ± | 2 | 34 % | 6 |
| As | 0.4 | ± | 0.1 | 38 % | 6 |
| Ni | 2.4 | ± | 0.7 | 30 % | 6 |

**Duisburg**

| deposition, µg/m²/day | | | | | |
|---|---|---|---|---|---|
| | Avg | | SD | rel SD | nr |
| Cd | 0.80 | ± | 0.06 | 7 % | 6 |
| Pb | 52 | ± | 2 | 4 % | 6 |
| As | 1.5 | ± | 0.1 | 10 % | 6 |
| Ni | 18 | ± | 1 | 8 % | 6 |

The uncertainty in the Bergerhoff measurements are based on only 6 parallel samples and is therefore not very robust, but its is clear that that samples are representative and reproducible. The uncertainties at Birkenes are relatively high due to very large precipitation in some periods that overflowed several of the samplers. The standard deviation is lowest at Duisburg due to higher pollution level, which is easier to measure.

Except that the between sampler uncertainty tends to be lower at the more polluted sites, but not always, there is no clear difference between the sites. Some elements have higher uncertainty at some sites while others not. This can be due to contamination and influence of local sources at the sites or issues regarding the laboratory. The range of standard deviation measured with parallel sampling is summarized in **Table 9**.

**Table 9.** Summary of the range of standard deviation for the different samplers, sites and elements.

| | Wet only collector | Bulk collector | Bergerhoff collector |
|---|---|---|---|
| Cd | 9 – 18 % | 11 – 32 % | 7 – 28 % |
| Pb | 7 – 15 % | 12 – 20 % | 4 – 44 % |
| As | 6 – 19 % | 9 – 22 % | 9 – 38% |
| Ni | 11 – 20 % | 12 – 48 % | 8 – 30 % |
| mm | 3 – 10% | 2 – 10 % | |

The precision in the wet only measurements is better than in the two other types of collectors. The open bucket collectors are influence by dry deposition, which creates an additional uncertainty in these measurements compared to the wet only collectors. The magnitude of dry deposition might not be equal between two neighbouring collectors due to different micrometeorological conditions. The added uncertainty in the funnel wash is also a contribution component. The bulk collector seems more precise than the Bergerhoff at low concentration levels.

### 5.3 Between laboratory uncertainty

The samples were split between laboratories as defined in Chapter 4. For the Bergerhoff sampler, three samplers were sent to one lab while one to the other. The laboratories did their own digestion procedures etc. For the wet only and bulk samplers, the procedure changed throughout the period. At Duisburg and Peyrusse all the samplers were digested and poured into smaller containers before a subset of the sample was sent to the other laboratory. In these exercises the uncertainty is then only representative of the analytical procedure + transport of the subset. At Copenhagen and Birkenes the parallel samplers was sent directly from the field sites to the two different laboratories for analysis. Here the uncertainty, in addition to the analytical measurements, included

uncertainty caused by sampling, transport and digestion procedures. In **Table 10** to **Table 12** the results from the between laboratory intercomparison are presented.

**Table 10.** Between laboratory uncertainties for the bulk measurements.

| Birkenes (UBA -NILU) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.131 ± | 0.013 | 10 % | 9 |
| Pb | 5.6 ± | 0.6 | 11 % | 9 |
| As | 0.56 ± | 0.06 | 12 % | 9 |
| Ni | 1.7 ± | 0.3 | 18 % | 9 |

| Peyrusse (UBA -NILU) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.085 ± | 0.004 | 4.4 % | 8 |
| Pb | 3.8 ± | 0.6 | 15 % | 8 |
| As | 0.40 ± | 0.04 | 10 % | 8 |
| Ni | 2.9 ± | 0.3 | 9.0 % | 8 |

| Copenhagen (UBA -FMI) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.062 ± | 0.012 | 20 % | 12 |
| Pb | 2.56 ± | 0.23 | 9 % | 12 |
| As | 0.25 ± | 0.048 | 19 % | 12 |
| Ni | 1.5 ± | 0.2 | 10 % | 12 |

| Duisburg (UBA -FMI) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.85 ± | 0.09 | 11 % | 5 |
| Pb | 48 ± | 6 | 13 % | 5 |
| As | 1.34 ± | 0.06 | 5 % | 5 |
| Ni | 7.7 ± | 0.6 | 8 % | 5 |

**Table 11.** Between laboratory uncertainties for the wet only measurements.

| Birkenes (UBA -NILU) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.105 ± | 0.004 | 4 % | 8 |
| Pb | 4.7 ± | 0.2 | 3 % | 8 |
| As | 0.38 ± | 0.03 | 8 % | 8 |
| Ni | 1.4 ± | 0.4 | 27 % | 8 |

| Peyrusse (UBA -NILU) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.102 ± | 0.007 | 7 % | 4 |
| Pb | 4.6 ± | 0.7 | 15 % | 4 |
| As | 0.37 ± | 0.04 | 12 % | 4 |
| Ni | 2.4 ± | 1.3 | 54 % | 4 |

| Copenhagen (UBA -FMI) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.053 ± | 0.005 | 10 % | 14 |
| Pb | 2.7 ± | 0.3 | 11 % | 14 |
| As | 0.22 ± | 0.04 | 18 % | 14 |
| Ni | 1.4 ± | 0.4 | 30 % | 13 |

| Duisburg (UBA -FMI) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.75 ± | 0.06 | 8 % | 5 |
| Pb | 48 ± | 4 | 7 % | 5 |
| As | 1.4 ± | 0.1 | 10 % | 5 |
| Ni | 6.3 ± | 1.3 | 21 % | 5 |

**Table 12.** Between laboratory uncertainties for the Bergerhoff measurements.

| Birkenes (EMD -LUA) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.1 ± | 0.3 | 316 % | 4 |
| Pb | 2.1 ± | 0.3 | 14 % | 4 |
| As | 0.3 ± | 0.4 | 136 % | 4 |
| Ni | 1.1 ± | 0.6 | 58 % | 4 |

| Peyrusse (LUA-EMD) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.06 ± | 0.11 | 186 % | 5 |
| Pb | 3 ± | 2 | 73 % | 6 |
| As | 0.29 ± | 0.20 | 69 % | 6 |
| Ni | 1.3 ± | 1.0 | 76 % | 6 |

| Copenhagen (LUA - EMD) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.11 ± | 0.01 | 10 % | 4 |
| Pb | 6 ± | 2 | 40 % | 4 |
| As | 0.4 ± | 0.3 | 61 % | 4 |
| Ni | 2.1 ± | 1.2 | 54 % | 4 |

| Duisburg (EMD - LUA) | deposition, µg/m²/day | | | |
|---|---|---|---|---|
| | Avg | SD rel SD | | nr |
| Cd | 0.80 ± | 0.13 | 16 % | 6 |
| Pb | 49.1 ± | 5.2 | 11 % | 6 |
| As | 1.42 ± | 0.223 | 16 % | 6 |
| Ni | 18 ± | 2 | 9 % | 6 |

The standard deviation between the laboratories for the wet only and bulk measurements are generally below 15% with some exceptions, especially for Ni. This is higher deviation than experienced in the laboratory intercomparison described in Chapter 3. But split samples are taken from unfiltrated samples which might be inhomogeneous, causing an inherit difference between the samples. For the Bergerhoff the uncertainty is generally much higher for the low concentration samples, but similar level as for the other methods at the polluted site in Duisburg. One should however notice that the estimates are based on very few samples and outliers have large effect. The laboratories use different analytical techniques, LUA with AAS while EMD use ICP-MS, and this may cause part of the uncertainty for the Bergerhoff sampling for the rural sites. In addition the comparison is done from an average of three samples from one lab with one for the second lab. At Birkenes there, in addition, were problem with overflow of the collector and snow clogging causing possible non-representative sampling. When looking at the Duisburg intercomparison in **Table 12** where both analytical methods perform well, the relative deviation is on the same level as the solid samples distributed in the laboratory intercomparison, see **Table 4**. This suggests that the main uncertainty in the Bergerhoff method for polluted areas is the sample preparation, including digestion and analysis.

It does not seem to be any difference between the wet only and bulk sample intercomparison either splitting the samples in the laboratory (Duisburg and Peyrusse) or in the field (Copenhagen and Birkenes). This indicate that the transport and sample preparation cause minor uncertainty in the total budget. But since the deviations are

higher than for the laboratory intercomparison, see **Table 3**. This indicates that the sampling method has large impact on the overall uncertainty.

### 5.4 Uncertainty in wet deposition and "total" deposition

The wet deposition can be measured using wet only or bulk collector. The bulk collector is however influenced by dry deposition; the magnitude of this depends on the sites. The precipitations collected from these two different collectors are compared to evaluate which sampler could be used for wet deposition at the different sites. The average of the two parallel collectors is compared with the average of the two parallel measurements of the other collector, see **Table 13**. The standard deviations are calculated using equation (1).

**Table 13.** Wet deposition from wet only and bulk collector, μg/($m^2$ day).

| Birkenes, wet only - bulk | | | | | | | |
|---|---|---|---|---|---|---|---|
| | WO | Bulk | Avg | | SD | rel SD | nr |
| Cd | 0.11 | 0.10 | 0.10 | ± | 0.02 | 20 % | 17 |
| Pb | 4.2 | 4.4 | 4.3 | ± | 0.7 | 15 % | 17 |
| As | 0.4 | 0.4 | 0.4 | ± | 0.1 | 13 % | 17 |
| Ni | 1.6 | 1.4 | 1.5 | ± | 0.6 | 43 % | 16 |

| Peyrusse, wet only - bulk | | | | | | | |
|---|---|---|---|---|---|---|---|
| | WO | Bulk | Avg | | SD | rel SD | nr |
| Cd | 0.05 | 0.07 | 0.06 | ± | 0.03 | 51 % | 16 |
| Pb | 2.2 | 2.5 | 2.4 | ± | 0.9 | 36 % | 16 |
| As | 0.19 | 0.24 | 0.21 | ± | 0.09 | 40 % | 16 |
| Ni | 0.7 | 1.4 | 1.0 | ± | 0.9 | 82 % | 16 |

| Copenhagen, wet only - bulk | | | | | | | |
|---|---|---|---|---|---|---|---|
| | WO | Bulk | Avg | | SD | rel SD | nr |
| Cd | 0.07 | 0.07 | 0.07 | ± | 0.02 | 26 % | 17 |
| Pb | 3.0 | 3.2 | 3.1 | ± | 1.1 | 35 % | 17 |
| As | 0.26 | 0.24 | 0.25 | ± | 0.07 | 28 % | 17 |
| Ni | 1.2 | 1.0 | 1.1 | ± | 0.5 | 41 % | 17 |

| Duisburg, wet only - bulk | | | | | | | |
|---|---|---|---|---|---|---|---|
| | WO | Bulk | Avg | | SD | rel SD | nr |
| Cd | 0.4 | 0.6 | 0.5 | ± | 0.2 | 30 % | 19 |
| Pb | 28 | 39 | 33 | ± | 16 | 49 % | 19 |
| As | 0.7 | 0.9 | 0.8 | ± | 0.2 | 23 % | 19 |
| Ni | 4 | 6 | 5 | ± | 2 | 41 % | 19 |

As was also seen in **Figure 6**, the average deposition for the two samplers is very similar at Birkenes and in Copenhagen. Duisburg and Peyrusse are more influenced by dry deposition causing an enhanced level in the bulk collector. The standard deviation is naturally also higher at these two sites. At Birkenes and at Copenhagen, the dry deposition has little influence; for some elements the average deposition load is even higher for the bulk collector. The standard deviation is between 13 % - 43 % at these two sites. The highest deviations are caused by a few episodes that don't have too strong impacts on the deposition load.

"Total" deposition as defined as wet deposition plus the dry deposition of particles to the open face collectors can be compared using Bergerhoff sampler and the bulk samplers + the amount deposited on the funnel wall, see
**Table 14**. One should notice that the real total deposition is usually higher since these collectors are not efficient for collecting small particles. The three Bergerhoff samples analysed at one lab are averaged and compared to the average of the two parallel bulk samples + the monthly funnel wash. In Peyrusse the funnel wash was only done the three first month, **Table 6**. But to get an estimate of the bulk + funnel for the whole period the average of funnel deposition from the first three months were used as an estimate in the months missing samples. This gives probably not a very big error since the funnel deposition contributes relatively little to the deposition in Peyrusse compared to the bulk precipitation. However the precision in the bulk + funnel compared to the Bergerhoff deposition is lowered due to this assumption. An additional uncertainty at

Peyrusse was that the bulk samples were not sampled weekly in June but one period that lasted for 35 days.

**Table 14.** Comparison of deposition using Bergerhoff (BH) and Bulk + funnel wash (B+F), *µg/(m² day).*

| Birkenes, Bergerhoff - bulk + funnel | | | | | | | |
|---|---|---|---|---|---|---|---|
| | B+F | BH | Avg | | SD | rel SD | nr |
| Cd | 0.10 | 0.10 | 0.10 | ± | 0.03 | 32 % | 6 |
| Pb | 4 | 2 | 3 | ± | 2 | 66 % | 6 |
| As | 0.4 | 0.3 | 0.4 | ± | 0.1 | 37 % | 6 |
| Ni | 1.4 | 1.1 | 1.2 | ± | 0.5 | 40 % | 6 |

| Peyrusse, Bergerhoff - bulk + funnel | | | | | | | |
|---|---|---|---|---|---|---|---|
| | B+F | BH | Avg | | SD | rel SD | nr |
| Cd | 0.09 | 0.06 | 0.08 | ± | 0.03 | 44 % | 5 |
| Pb | 3 | 2 | 3 | ± | 1 | 35 % | 5 |
| As | 0.30 | 0.32 | 0.31 | ± | 0.05 | 15 % | 5 |
| Ni | 1.9 | 1.1 | 1.5 | ± | 0.7 | 49 % | 5 |

| Copenhagen, Bergerhoff - bulk + funnel | | | | | | | |
|---|---|---|---|---|---|---|---|
| | B+F | BH | Avg | | SD | rel SD | nr |
| Cd | 0.07 | 0.11 | 0.09 | ± | 0.03 | 31 % | 6 |
| Pb | 4 | 7 | 5 | ± | 3 | 48 % | 6 |
| As | 0.2 | 0.4 | 0.3 | ± | 0.1 | 40 % | 6 |
| Ni | 1 | 2 | 2 | ± | 1 | 65 % | 6 |

| Duisburg, Bergerhoff - bulk + funnel | | | | | | | |
|---|---|---|---|---|---|---|---|
| | B+F | BH | Avg | | SD | rel SD | nr |
| Cd | 0.9 | 0.8 | 0.9 | ± | 0.2 | 21 % | 6 |
| Pb | 69 | 52 | 60 | ± | 15 | 26 % | 6 |
| As | 1.3 | 1.5 | 1.4 | ± | 0.2 | 17 % | 6 |
| Ni | 10 | 18 | 14 | ± | 6 | 46 % | 6 |

Considering the very different sampling procedures and the few comparison points the difference between the two approaches is quite small at the three sites with low deposition level. The difference is bigger at Duisburg even though the relative standard deviation is low. Nickel is in general higher compared the Bergerhoff analysis by LUA and EMD compared to the bulk + funnel analysed at FMI and NILU. The reason for this is due to the different work up of the samples. NILU and FMI analysed the collected water (including acid washing of the funnels) while LUA and EMD evaporated the water and digested the residue with the microwave using $H_2O_2/HNO_3$ as reagent. It is known that a great fraction of nickel is bound to large particles as silicate or oxide and this is not detected without a complete digestion of the sample. It is therefore expected that, especially for Ni, the level should be higher in the Bergerhoff samplers. This is not the case at Birkenes and Peyrusse. At Birkenes, this can be due to the fact that it was overflow and snow clogging in the Bergerhoff collectors, which probably underestimate the deposition load.

## 5.5 Testing for the efficiency of funnel rinsing

The rinsing effectiveness between samples was tested at the Duisburg site. The funnel was cleaned with acidic water every week. To test if this was sufficient to remove all the particles from the funnel wall a second rinsing was performed. The funnel was cleaned by pouring twice 250 ml of ultrapure 1 % nitric acid solution along the walls of the funnel. The two rinsing solutions were collected into separate bottles for further laboratory analysis. The results are shown in **Figure 7**. The 1st rinsing was found to be very effective as the solution contained 85 % - 90 % of total cadmium and lead and 76 % - 84 % of total nickel and arsenic. For all the elements the results from parallel collectors were very reproducible. This means that the efficiency of the first rinsing is satisfactory.

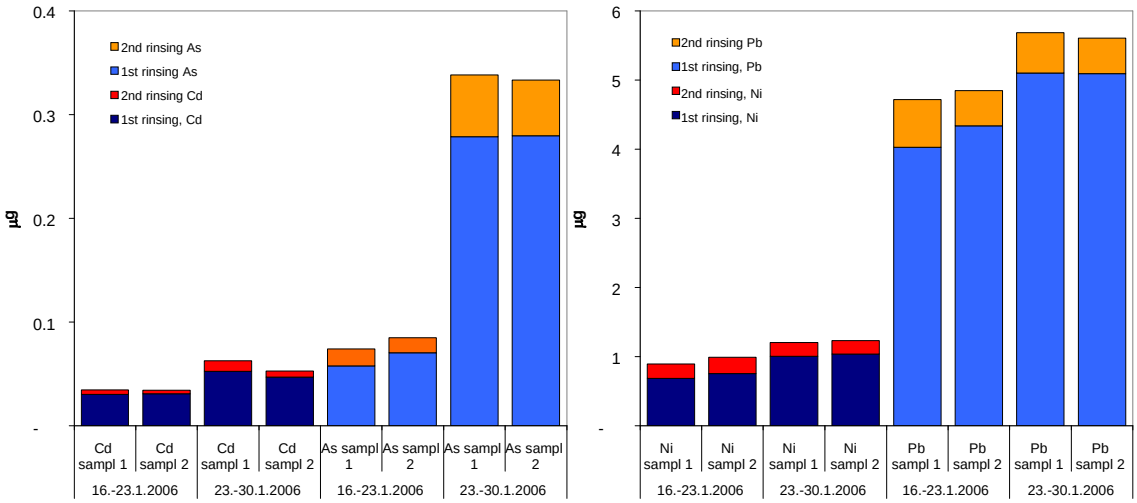


**Figure 7.** Rinsing efficiency of bulk collector funnels.

## 5.6 Analyse the effect of unaccounted dry deposition in collector bottle

In dry periods some deposition might be deposited in the collection bottle, which is not accounted for, either because of too little precipitation or no precipitation at all. The heavy metals deposited in the collector can either be from precipitation where some of the volume might have evaporated or from dry deposition. To estimate how important this might be, the dry deposition was analysed from the bottle after dry periods in Copenhagen and Duisburg. In the entire field experiment there were altogether three weeks with no precipitation and four weeks with very small precipitation amount (< 0.6 mm). In order to analyse the dry deposition, a small amount (20 ml) of ultrapure 1 % nitric acid was added into the collector bottle in the laboratory. The depositions found in parallel collectors are very equal (there is one odd result from German site). The depositions are higher, especially nickel and lead, at Duisburg site than at Copenhagen site. However when comparing with the total bulk deposition at both sites the unaccounted deposition caused by this effect is insignificant, less than 0.5 % for all elements at both sites, see **Table 15**, meaning that this has not been a source of large error in the estimates in the average deposition load during the field trials.

**Table 15.** Estimate of the importance of dry deposition in the bulk collector during dry periods.

| Copenhagen, µg/(m²day) | | | | | |
|---|---|---|---|---|---|
| Period | | Cd | As | Ni | Pb |
| 12.19.09.2006 | Sampler 1 | 0.002 | 0.009 | 0.04 | 0.12 |
| | Sampler 2 | 0.002 | 0.009 | 0.03 | 0.13 |
| 19-26.09.2006 | Sampler 1 | n.d. | n.d. | 0.01 | 0.05 |
| | Sampler 2 | 0.0001 | n.d. | 0.01 | 0.06 |
| 10-17.10.2006 | Sampler 1 | 0.000 | 0.002 | 0.08 | 0.13 |
| | Sampler 2 | 0.001 | 0.001 | 0.03 | 0.07 |
| 19-26.12.2006 | Sampler 1 | 0.003 | 0.011 | 0.12 | 0.07 |
| | Sampler 2 | 0.003 | 0.009 | 0.14 | 0.11 |
| Average dry in collector, µg/(m² day) | | 0.002 | 0.007 | 0.06 | 0.09 |
| Total unaccounted dep, µg/m² | | 0.03 | 0.1 | 0.8 | 2 |
| Total bulk dep in field trial, µg/m² | | 11 | 37 | 151 | 501 |
| Per cent unaccounted deposition | | 0.2 % | 0.3 % | 0.5 % | 0.4 % |

| Duisburg, µg/(m² day) | | | | | |
|---|---|---|---|---|---|
| Period | | Cd | As | Ni | Pb |
| 02 -09.05.2006 | Sampler 1 | 0.017 | 0.03 | 0.17 | 2.07 |
| | Sampler 2 | 0.011 | 0.02 | 0.15 | 1.33 |
| 06-13.06.2006 | Sampler 1 | 0.0002 | 0.001 | 0.01 | 0.01 |
| | Sampler 2 | 0.001 | 0.002 | 0.01 | 0.02 |
| 27.06.-04.07.2006 | Sampler 1 | 0.023 | 0.01 | 0.16 | 2.29 |
| Average dry in collector, µg/(m² day) | | 0.01 | 0.01 | 0.11 | 1.3 |
| Total unaccounted dep, µg/m² | | 0.3 | 0.3 | 2.3 | 28.0 |
| Total bulk dep in field trial, µg/m² | | 96 | 138 | 982 | 6090 |
| Per cent unaccounted deposition | | 0.3 % | 0.2 % | 0.2 % | 0.5 % |

## 5.7 Analyse the loss of heavy metals in the filtration of the sample

Acidified precipitation samples, especially those collected at urban and industrial sites or at remote sites in Southern and Central Europe, may still contain non-dissolved material. Such non-homogenous samples shall be filtered before analysis. However, possibly non-dissolved metals will not be detected, if only the filtrate is analysed. Therefore, the guidance document for the field test (CEN/TC264/WG20 N025) foresees a special test to check if there is evidence for non-dissolved metals in the acidified samples. The filtration test is based on the digestion and analysis of sub-samples.

Unfortunately, it is not possible to take representative sub-samples of non-homogenous precipitation samples. Thus this test will only indicate the existence/ occurrence of non-dissolved metals in the samples but will not allow quantifying the non-dissolved metal content. Two labs performed the test, UBA and FMI. UBA analyses wet-only samples, FMI analyses bulk samples.

For filtration tests, equipment for vacuum filtration is used: About 20 - 50 ml of 1% nitric acid is filtered to rinse the filter (cellulose acetate filter with 0.45 µm pore size) and the filtrate is discarded. Then exactly 100 ml of the well-mixed sample is filtered into an acid cleaned PFA bottle for ICP/MS analysis. The filter is digested following the procedure given in EN14902 with some minor modifications.

The total metal content in the precipitation samples is calculated by adding the metal content found in the filtrate (dissolved metal) and the metal content found in the filter residue (non-dissolved metal). The percentages of dissolved and non-dissolved metals in precipitation (collected in sampling bottle) and funnel rinsing are shown in **Figure 8**. Notice that only a limited number of sub-samples ($2 \leq n \leq 8$) is analysed.

These figures show that – for the wet-only collectors - there is some evidence of non-dissolved metals in precipitation samples (collected in the sampling bottle) as well as in the funnel rinsing solutions. The importance of the non-dissolved metal fraction is different for different sites. It is more important for the industrial site Duisburg and the southern remote site Peyrusse. The importance of non-dissolved metal is most important for nickel. However, there is no such evidence for the bulk collectors. A possible explanation might be that it is not possible to take representative sub-samples of non-homogenous precipitation samples, but there are not enough samples for bulk collectors to conclude anything.

Despite any restrictions of this filtration test, e.g. limited number of samples analysed; possibly non-representative sub-samples; no quantification of the non-dissolved metal content in the complete sample, there is some evidence for non-dissolved metals in precipitation samples. This type of filtration test could be useful to estimate the importance of non-dissolved metals in precipitation samples at a special site. Checking for non-dissolved metals in precipitation samples would be especially important when a new station is installed.

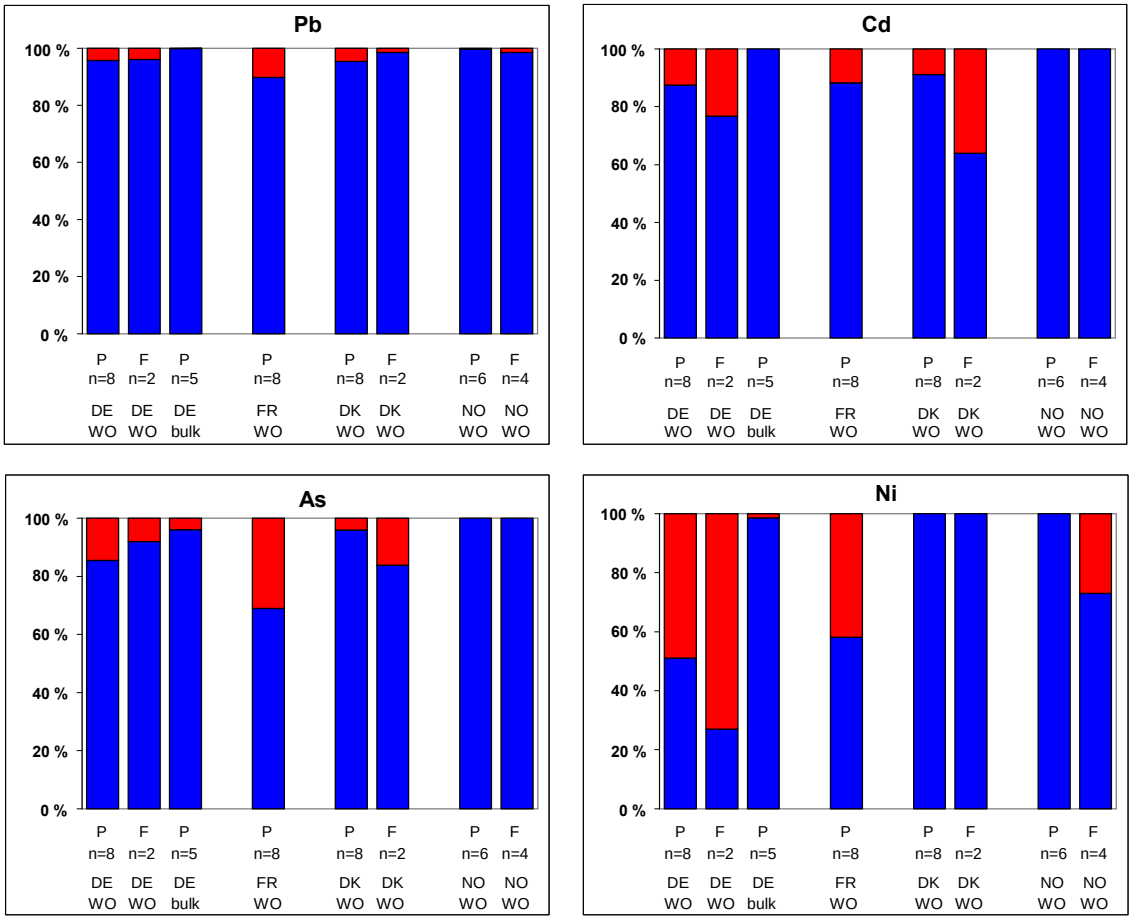


**Figure 8.** Percentage of dissolved (blue) and non-dissolved (red) metal. Median of n samples. DE: Duisburg, FR: Peyrusse, DK: Copenhagen, NO: Brkenes, WO: wet only.

# 6 Conclusion

As have been demonstrated, the different sampling and analytical methods can under some conditions be comparable, while in others not. It is quite clear that different sampling strategy is needed for rural and industrial sites. So the conclusions on uncertainties and sample approach are made differently for industrial and rural sites. For the urban site in Copenhagen the results were very similar to the rural sites. Other urban environment may be more polluted, and if so should follow the recommendations for the industrial area.

**Industrial or high-polluted region**

At highly polluted sites with much dust, the wet only sampler will not be able to sample all the deposition, but the sampler is excellent to determine wet deposition. Both AAS and ICP MS work well with highly polluted samples. If bulk is used, it is necessary to analyse the deposition on the funnel walls to estimate the importance of dry deposition. In addition one should do a filtration test to estimate the importance of non-dissolved metals in precipitation samples. Furthermore, it is often necessary to use a digestion technique enabling to dissolve all the heavy metal oxides.

If following these recommendations, the uncertainty can be estimated looking at the Duisburg intercomparison. The comparison between bulk+funnel and Bergerhoff gives a total picture of the uncertainty. It included methodology differences, analytical procedures, transport etc. For Ni the comparison is not valid due to the reason mentioned above, but for the three other elements, the standard deviation is 26% or less, see
**Table 14**. It is reasonable to believe that the uncertainty in Ni is in the same order when looking at the results in the between sampler uncertainties, see **Table 12**. This means that the expanded uncertainty at 95 % confidence level can be estimated to be 52 %:

| | |
|---|---|
| Standard deviation | u < 26 % |
| Expanded uncertainty: | U = k*u: 2* 26% = 52 % |

This is well within the data quality objectives given in the 4$^{th}$ daughter directive.

**Rural region**

In regions with much rain and/or snow the Bergerhoff collector do not work properly. It has problem to collect all the rain for monthly sampling, and for snow the opening diameter is too small and the collector volume too little to collect the snow efficiency; this collector will thus underestimate the deposition. Only ICP MS is proven to work well at low concentration samples. Wet only collector as being the reference method in EMEP has proven to be better than the required 30 % uncertainty in annual average at rural sites, see **Table 7**.

At sites with frequent rain the deposition is covered by wet deposition mainly, and bulk and wet only work equally well. The difference between bulk and wet only sampling at Birkenes and Copenhagen can be used as representative uncertainty for the deposition at background sites. The standard deviations are below 35 % for all elements except Ni, see **Table 13**. As mentioned earlier, Ni is more difficult since it is easier to contaminate the sample as well as digestion issues. But it is not likely that the higher standard

deviation for Ni is due to different method because the average deposition is higher in the wet only collector, it would be expected to be the opposite if it was due to methodology difference since the bulk collector collects some dry deposition. In addition all these laboratories use ICP MS and same digestion procedure. But as also seen from the between lab comparison (**Table 10** and **Table 11**), the relative standard deviation can be quite high for Ni indicating that there might be difficulties with contamination. However the relative difference for the four elements are similar for the bulk deposition (**Table 6**) in Copenhagen as well as the wet only deposition (**Table 7**) in both Copenhagen and Birkenes. But, it is relatively high standard deviation for Ni at Birkenes for the bulk collector measurements, 48 % **Table 6**. In the statistic evaluation, outliers have not been deleted and the deviations are therefore higher than one should expect than only considering the methodology. One should therefore look at the standard deviation at the 3 other elements only, since these are less influenced by contamination. If so one can conclude that both bulk and wet only are within the data quality objectives given in the 4$^{th}$ DD. The expanded uncertainty at 95 % confidence level for rural areas with frequent rain events can be estimated to be 70 %:

| | |
|---|---|
| Standard deviation | u < 35 % |
| Expanded uncertainty: | U = k*u: 2* 35 % = 70 % |

At drier rural sites, wet only is the only collector giving correct estimate of the wet deposition, which is required by the EMEP programme. At Peyrusse, the influence on dry deposition on the bulk sampling makes a bias compared with wet only showing that these two methods don't measure the same thing. To estimate the standard deviation of the wet deposition one can look at the parallel wet only collectors at Peyrusse, **Table 7**. The standard deviation is below 15 % for all the elements meaning that the uncertainty at the 95% confidence level is 30 %.

If it is required to estimate the influence of dry deposition either Bergerhoff or bulk + funnel should be used. This comparison at Peyrusse, however gives high relative standard deviation, about 50 %, but this calculation is based on only 5 parallel measurements. Furthermore, the two last months there was no funnel wash; the deposition on the funnel was estimated using the average from the three first months. One could use only three months of data, but that is too few parallel samples to quantify the uncertainty. If looking at the parallel Bergerhoff samples, see **Table 8**, the standard deviation is below 12 % while for bulk it is below 32 %, see **Table 6**, meaning that both these methods individually have an expanded uncertainty below the data quality objectives of 70 %.

To conclude, the standard method for the determination of Pb, Cd, As and Ni in depositions being developed by the CEN/TC 264/WG 20 fulfil the data quality objectives of 70 % uncertainty at a 95 % confidence level. But not all collectors are suited to be used at all the different type of sites. Bergerhoff or bulk + funnel should be used at industrial sites while bulk or wet only at rural sites.

# 7 Bibliography


1 Manual for sampling and chemical analysis. Revised November 2001. Kjeller, Norwegian Institute for Air Research (EMEP/CCC-Report 1/95). URL: http://www.nilu.no/projects/ccc/manual/index.html.

2 The Fourth Daughter Directive. Directive 2004/107/EC of the European Parliament and of the council of 15th December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air.

3 The First Daughter Directive. Council directive 1999/30/EC of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air.

4 EN-ISO 20988:2007 Air quality - Guidelines for estimating measurement uncertainty (ISO 20988:2007)

5 EN 14902:2005 Ambient air quality - Standard method for the measurement of Pb, Cd, As and Ni in the PM10 fraction of suspended particulate matter

6 CEN/TC 264/WG 20 N025 Guide on Execution of preliminary field tests and ield tests - CEN/TC264/WG 20 Deposition of heavy metals except Hg

## Appendix. The raw data from the field trials.

# A.1 Results from split samples from the field trials in Peyrusse

**Split bulk samples analysed at NILU and UBA**

| | | | | | concentrations, µg/l | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| first day | last day | days | collector | mm | Pb UBA | Pb NILU | Cd UBA | Cd NILU | As UBA | As NILU | Ni UBA | Ni NILU | Pb UBA | Pb NILU | Cd UBA | Cd NILU | As UBA | As NILU | Ni UBA | Ni NILU |
| 13.06.06 | 20.06.06 | 7 | E | 38 | 1.28 | 1.54 | 0.03 | 0.03 | 0.15 | 0.14 | 0.69 | 0.59 | 7.01 | 8.46 | 0.14 | 0.14 | 0.83 | 0.78 | 3.80 | 3.21 |
| 13.06.06 | 20.06.06 | 7 | F | 38 | 1.48 | 1.35 | 0.03 | 0.02 | 0.16 | 0.13 | 0.58 | 0.56 | 8.13 | 7.39 | 0.15 | 0.13 | 0.86 | 0.73 | 3.16 | 3.06 |
| 20.06.06 | 28.06.06 | 8 | E | 16 | 1.64 | 1.57 | 0.05 | 0.05 | 0.23 | 0.21 | 1.03 | 1.18 | 3.26 | 3.11 | 0.10 | 0.10 | 0.45 | 0.42 | 2.05 | 2.34 |
| 20.06.06 | 28.06.06 | 8 | F | 8 | 3.06 | 1.83 | 0.15 | 0.15 | 0.29 | 0.25 | 1.74 | 1.74 | 3.18 | 1.91 | 0.16 | 0.16 | 0.30 | 0.26 | 1.80 | 1.81 |
| 08.08.06 | 16.08.06 | 8 | E | 5 | 2.54 | 2.94 | 0.05 | 0.06 | 0.29 | 0.30 | 2.03 | 2.52 | 1.48 | 1.71 | 0.03 | 0.03 | 0.17 | 0.17 | 1.18 | 1.46 |
| 08.08.06 | 16.08.06 | 8 | F | 5 | 5.21 | 5.44 | 0.04 | 0.04 | 0.36 | 0.41 | 5.34 | 6.12 | 3.03 | 3.16 | 0.02 | 0.03 | 0.21 | 0.24 | 3.11 | 3.56 |
| 16.08.06 | 22.08.06 | 6 | E | 18 | 0.77 | 0.62 | 0.01 | 0.02 | 0.08 | 0.10 | 1.41 | 1.57 | 2.30 | 1.86 | 0.04 | 0.05 | 0.23 | 0.29 | 4.24 | 4.71 |
| 16.08.06 | 22.08.06 | 6 | F | 22 | 0.58 | 0.76 | 0.01 | 0.01 | 0.07 | 0.07 | 0.90 | 1.01 | 2.14 | 2.83 | 0.05 | 0.05 | 0.27 | 0.25 | 3.34 | 3.73 |

**Split wet only samples analysed at NILU and UBA**

| | | | | | concentrations, µg/l | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| first day | last day | days | collector | mm | Pb NILU | Pb UBA | Cd NILU | Cd UBA | As NILU | As UBA | Ni NILU | Ni UBA | Pb NILU | Pb UBA | Cd NILU | Cd UBA | As NILU | As UBA | Ni NILU | Ni UBA |
| 13.06.2006 | 20.06.2006 | 7 | G | 42.9 | 1.32 | 1.12 | 0.03 | 0.03 | 0.11 | 0.10 | 0.82 | 0.42 | 8.06 | 6.87 | 0.20 | 0.19 | 0.67 | 0.59 | 5.06 | 2.60 |
| 20.06.2006 | 26.06.2006 | 6 | G | 20 | 2.22 | 1.80 | 0.04 | 0.04 | 0.17 | 0.15 | 1.35 | 0.52 | 7.41 | 5.99 | 0.15 | 0.13 | 0.58 | 0.50 | 4.49 | 1.75 |
| 18.07.2006 | 25.07.2006 | 7 | G | 16.8 | 1.50 | 1.23 | 0.02 | 0.02 | 0.10 | 0.08 | 0.63 | 0.42 | 3.60 | 2.94 | 0.06 | 0.05 | 0.24 | 0.20 | 1.50 | 1.01 |
| 25.07.2006 | 01.08.2006 | 7 | G | 8.6 | 0.95 | 0.81 | 0.01 | 0.01 | 0.07 | 0.06 | 1.38 | 1.11 | 1.16 | 0.99 | 0.02 | 0.01 | 0.09 | 0.07 | 1.70 | 1.37 |

**Parallell Bergerhoff samples analysed at EMD and LUA**

| | | | AVG 3 samples EMD | | | | LUA, one sample | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| From | To | days | As | Ni | Cd | Pb | As | Ni | Cd | Pb |
| 7-mar-06 | 4-apr-06 | 28 | 0.24 | 0.70 | 0.06 | 2.31 | **0.25** | 1.14 | 0.07 | 3.81 |
| 4-apr-06 | 2-mai-06 | 28 | 0.25 | 0.94 | 0.06 | 1.96 | 0.18 | 1.78 | 0.06 | **3.62** |
| 2-mai-06 | 30-mai-06 | 28 | 0.24 | 0.86 | 0.06 | 2.26 | 0.25 | 1.72 | | 6.45 |
| 30-mai-06 | 28-jun-06 | 29 | 0.70 | 2.56 | 0.09 | 3.86 | 0.81 | 2.45 | 0.12 | 7.76 |
| 28-jun-06 | 25-jul-06 | 27 | 0.16 | 0.58 | 0.04 | 1.71 | 0.12 | 1.42 | 0.04 | 2.21 |
| 25-jul-06 | 22-aug-06 | 28 | 0.19 | 0.46 | 0.02 | 1.54 | 0.12 | 0.74 | 0.03 | 2.63 |

# A.2 Results from split samples from the field trials in Copenhagen

**Split bulk samples analysed at FMI and UBA**

| | | | | | | concentrations, µg/l | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| first day | last day | days | collector | mm | mm avg | Pb UBA | Pb FMI | Cd UBA | Cd FMI | As UBA | As FMI | Ni UBA | Ni FMI | Pb UBA | Pb FMI | Cd UBA | Cd FMI | As UBA | As FMI | Ni UBA | Ni FMI |
| 01.11.06 | 07.11.06 | 6 | A | 8.36 | 8.36 | 1.35 | 0.98 | 0.04 | 0.02 | 0.16 | 0.13 | 0.40 | 0.47 | 1.88 | 1.36 | 0.06 | 0.03 | 0.22 | 0.18 | 0.56 | 0.66 |
| 07.11.06 | 15.11.06 | 8 | A | 33.05 | 33.05 | 1.38 | 1.21 | 0.03 | 0.02 | 0.11 | 0.07 | 0.27 | 0.20 | 5.69 | 5.01 | 0.14 | 0.09 | 0.44 | 0.30 | 1.14 | 0.83 |
| 15.11.06 | 21.11.06 | 6 | A | 16.09 | 16.28 | 1.61 | 1.58 | 0.04 | 0.03 | 0.17 | 0.14 | 1.46 | 1.39 | 4.32 | 4.24 | 0.11 | 0.09 | 0.46 | 0.38 | 3.92 | 3.72 |
| 15.11.06 | 21.11.06 | 6 | B | 16.47 | 16.28 | 1.66 | 1.87 | 0.03 | 0.03 | 0.16 | 0.16 | 1.20 | 1.30 | 4.54 | 5.13 | 0.08 | 0.09 | 0.43 | 0.44 | 3.29 | 3.58 |
| 21.11.06 | 28.11.06 | 7 | A | 11.64 | 11.72 | 1.26 | 1.33 | 0.03 | 0.03 | 0.10 | 0.07 | 1.31 | 1.30 | 2.10 | 2.21 | 0.05 | 0.05 | 0.16 | 0.12 | 2.18 | 2.16 |
| 21.11.06 | 28.11.06 | 7 | B | 11.79 | 11.72 | 0.91 | 1.01 | 0.03 | 0.03 | 0.09 | 0.08 | 1.20 | 1.29 | 1.54 | 1.69 | 0.05 | 0.05 | 0.15 | 0.14 | 2.03 | 2.18 |
| 28.11.06 | 05.12.06 | 7 | A | 9.63 | 9.63 | 1.72 | 1.79 | 0.04 | 0.04 | 0.13 | 0.12 | 0.52 | 0.38 | 2.37 | 2.46 | 0.06 | 0.06 | 0.18 | 0.17 | 0.72 | 0.53 |
| 28.11.06 | 05.12.06 | 7 | B | 9.64 | 9.63 | 1.59 | 1.79 | 0.04 | 0.04 | 0.14 | 0.11 | 0.49 | 0.54 | 2.18 | 2.47 | 0.06 | 0.06 | 0.19 | 0.15 | 0.67 | 0.74 |
| 16.01.07 | 24.01.07 | 8 | A | 42.97 | 43.34 | 0.41 | 0.43 | 0.01 | 0.01 | 0.07 | 0.05 | 0.26 | 0.21 | 2.18 | 2.31 | 0.07 | 0.05 | 0.37 | 0.26 | 1.42 | 1.14 |
| 16.01.07 | 24.01.07 | 8 | B | 43.72 | 43.34 | 0.40 | 0.41 | 0.01 | 0.01 | 0.06 | 0.04 | 0.24 | 0.23 | 2.19 | 2.24 | 0.06 | 0.06 | 0.35 | 0.23 | 1.33 | 1.28 |
| 24.01.07 | 30.01.07 | 6 | A | 13.82 | 13.76 | 0.32 | 0.31 | 0.02 | 0.02 | 0.08 | 0.06 | 0.24 | 0.42 | 0.73 | 0.71 | 0.05 | 0.04 | 0.18 | 0.14 | 0.55 | 0.96 |
| 24.01.07 | 30.01.07 | 6 | B | 13.70 | 13.76 | 0.39 | 0.48 | 0.01 | 0.01 | 0.07 | 0.07 | 0.20 | 0.25 | 0.89 | 1.10 | 0.03 | 0.03 | 0.16 | 0.16 | 0.46 | 0.57 |

**Split wet only samples analysed at FMI and UBA**

| | | | | | | concentrations, µg/l | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| first day | last day | days | collector | mm | mm avg | Pb FMI | Pb UBA | Cd FMI | Cd UBA | As FMI | As UBA | Ni FMI | Ni UBA | Pb FMI | Pb UBA | Cd FMI | Cd UBA | As FMI | As UBA | Ni FMI | Ni UBA |
| 01.11.06 | 07.11.06 | 6 | 1 | 8.09 | 8.09 | 0.82 | 0.72 | 0.01 | 0.01 | 0.11 | 0.12 | 0.18 | 0.32 | 1.10 | 0.97 | 0.01 | 0.02 | 0.15 | 0.16 | 0.25 | 0.43 |
| 01.11.06 | 07.11.06 | 6 | 2 | 8.35 | 8.35 | 1.16 | 1.16 | 0.01 | 0.02 | 0.09 | 0.12 | 0.50 | 0.68 | 1.62 | 1.62 | 0.02 | 0.02 | 0.12 | 0.16 | 0.70 | 0.94 |
| 07.11.06 | 15.11.06 | 8 | 1 | 33.66 | 33.66 | 1.80 | 1.54 | 0.02 | 0.02 | 0.08 | 0.09 | 0.02 | 0.31 | 7.57 | 6.47 | 0.10 | 0.10 | 0.35 | 0.40 | 0.10 | 1.31 |
| 07.11.06 | 15.11.06 | 8 | 2 | 34.09 | 34.09 | 1.64 | 1.57 | 0.02 | 0.02 | 0.06 | 0.09 | 0.02 | 0.37 | 7.00 | 6.71 | 0.08 | 0.10 | 0.25 | 0.40 | 0.08 | 1.59 |
| 15.11.06 | 21.11.06 | 6 | 1 | 16.20 | 16.20 | 1.61 | 1.34 | 0.03 | 0.03 | 0.15 | 0.14 | 0.69 | 0.48 | 4.36 | 3.61 | 0.09 | 0.08 | 0.39 | 0.39 | 1.88 | 1.28 |
| 15.11.06 | 21.11.06 | 6 | 2 | 16.30 | 16.30 | 1.45 | 1.37 | 0.03 | 0.03 | 0.13 | 0.15 | 0.56 | 0.70 | 3.94 | 3.71 | 0.08 | 0.07 | 0.34 | 0.41 | 1.52 | 1.91 |
| 21.11.06 | 28.11.06 | 7 | 1 | 11.25 | 11.25 | 1.17 | 1.07 | 0.03 | 0.03 | 0.07 | 0.08 | 1.31 | 1.23 | 1.89 | 1.72 | 0.05 | 0.05 | 0.12 | 0.13 | 2.11 | 1.98 |
| 21.11.06 | 28.11.06 | 7 | 2 | 11.13 | 11.13 | 1.23 | 1.08 | 0.04 | 0.04 | 0.08 | 0.09 | 1.50 | 1.43 | 1.95 | 1.71 | 0.06 | 0.06 | 0.12 | 0.14 | 2.38 | 2.27 |
| 28.11.06 | 05.12.06 | 7 | 1 | 9.74 | 9.74 | 0.95 | 0.83 | 0.03 | 0.03 | 0.07 | 0.09 | 5.59 | 0.61 | 1.32 | 1.16 | 0.05 | 0.04 | 0.10 | 0.13 | | |
| 28.11.06 | 05.12.06 | 7 | 2 | 9.41 | 9.41 | 1.06 | 1.02 | 0.03 | 0.03 | 0.07 | 0.09 | 0.53 | 0.61 | 1.42 | 1.36 | 0.05 | 0.04 | 0.10 | 0.13 | 0.71 | 0.81 |
| 16.01.07 | 24.01.07 | 8 | 1 | 43.24 | 43.24 | 0.51 | 0.43 | 0.01 | 0.01 | 0.05 | 0.05 | 0.22 | 0.23 | 2.75 | 2.32 | 0.05 | 0.05 | 0.26 | 0.27 | 1.21 | 1.22 |
| 16.01.07 | 24.01.07 | 8 | 2 | 43.51 | 43.51 | 0.43 | 0.43 | 0.01 | 0.01 | 0.04 | 0.05 | 0.28 | 0.35 | 2.35 | 2.34 | 0.05 | 0.05 | 0.20 | 0.26 | 1.51 | 1.92 |
| 24.01.07 | 30.01.07 | 6 | 1 | 12.00 | 12.00 | 0.52 | 0.46 | 0.01 | 0.01 | 0.07 | 0.08 | 1.52 | 1.51 | 1.04 | 0.93 | 0.02 | 0.03 | 0.13 | 0.16 | 3.03 | 3.01 |
| 24.01.07 | 30.01.07 | 6 | 2 | 12.45 | 12.45 | 0.53 | 0.57 | 0.01 | 0.01 | 0.04 | 0.08 | 0.36 | 0.35 | 1.09 | 1.18 | 0.02 | 0.03 | 0.09 | 0.17 | 0.75 | 0.74 |

**Parallell Bergerhoff samples analused at EMD and LUA**

| | | | AVG 3 samples LUA | | | | EMD, one sample | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| From | To | days | As | Ni | Cd | Pb | As | Ni | Cd | Pb | |
| 5-sep-06 | 3-okt-06 | 28 | 0.42 | 1.09 | 0.10 | 5.03 | | | | | |
| 3-okt-06 | 1-nov-06 | 29 | 0.47 | 1.88 | 0.11 | 5.86 | 0.572 | 1.882 | 0.11 | 8.20 | |
| 1-nov-06 | 28-nov-06 | 27 | 0.49 | 3.29 | 0.08 | 9.64 | 0.45 | 2.58 | 0.08 | 5.04 | |
| 28-nov-06 | 27-des-06 | 29 | 0.25 | 1.87 | 0.14 | 6.34 | 0.168 | 1.002 | 0.07 | 2.23 | not used since part of sample lost |
| 27-des-06 | 24-jan-07 | 28 | 0.25 | 2.68 | 0.11 | 10.22 | 0.406 | 1.616 | 0.11 | 3.14 | |
| 24-jan-07 | 20-feb-07 | 27 | 0.34 | 3.49 | 0.12 | 6.25 | 0.476 | 1.469 | 0.12 | 4.97 | |
| | | AVG | 0.37 | 2.38 | 0.11 | 7.23 | 0.48 | 1.89 | 0.11 | 5.34 | |

# A.3Results from split samples from the field trials in Duisburg

**Split bulk samples analysed at FMI and UBA**

| | | | concentrations, µg/l | | | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | mm | mm | | Pb | Pb | Cd | Cd | As | As | Ni | Ni | Pb | Pb | Cd | Cd | As | As | Ni | Ni |
| **first day** | **last day** | **days** | **UBA** | **FMI** | mm avg | UBA | **FMI** | UBA | **FMI** | UBA | **FMI** | UBA | **FMI** | UBA | **FMI** | UBA | **FMI** | UBA | **FMI** | UBA | **FMI** |
| 09.05.06 | 16.05.06 | 7 | 11.70 | 11.70 | 11.70 | 10.58 | 12.50 | 0.24 | 0.30 | 0.55 | 0.65 | 2.10 | 2.83 | 17.68 | 20.89 | 0.40 | 0.50 | 0.91 | 1.09 | 3.51 | 4.73 |
| 16.05.06 | 23.05.06 | 7 | 61.74 | 49.24 | 55.49 | 8.00 | 9.10 | 0.09 | 0.10 | 0.18 | 0.21 | 0.89 | 1.28 | 63.42 | 72.14 | 0.68 | 0.78 | 1.40 | 1.67 | 7.06 | 10.15 |
| 23.05.05 | 30.05.05 | 7 | 48.16 | 48.06 | 48.11 | 7.83 | 8.45 | 0.15 | 0.17 | 0.20 | 0.22 | 1.14 | 1.46 | 53.82 | 58.08 | 1.06 | 1.14 | 1.37 | 1.52 | 7.84 | 10.03 |
| 13.06.06 | 20.06.06 | 7 | 33.88 | 33.88 | 33.88 | 4.42 | 4.76 | 0.11 | 0.12 | 0.30 | 0.34 | 1.09 | 1.38 | 21.39 | 23.04 | 0.52 | 0.60 | 1.45 | 1.66 | 5.27 | 6.68 |
| 20.06.2006 | 27.06.2006 | 7 | 9.54 | 10.14 | 9.84 | 50.71 | 53.66 | 0.65 | 0.64 | 0.92 | 1.03 | 2.57 | 2.84 | 71.28 | 75.43 | 0.91 | 0.90 | 1.29 | 1.45 | 3.61 | 3.99 |

**Split wet only samples analysed at FMI and UBA**

| | | | concentrations, µg/l | | | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | mm | mm | | Pb | Pb | Cd | Cd | As | As | Ni | Ni | Pb | Pb | Cd | Cd | As | As | Ni | Ni |
| **first day** | **last day** | **days** | **UBA** | **FMI** | mm avg | **UBA** | FMI | **UBA** | FMI | **UBA** | FMI | **UBA** | FMI | UBA | FMI | UBA | FMI | UBA | FMI | UBA | FMI |
| 09.05.06 | 16.05.06 | 7 | 10.06 | 10.06 | 10.06 | 15.80 | 21.57 | 0.39 | 0.55 | 0.83 | 0.95 | 4.65 | 4.54 | 22.72 | 31.01 | 0.56 | 0.79 | 1.20 | 1.37 | 6.69 | 6.53 |
| 16.05.06 | 23.05.06 | 7 | 46.46 | 46.46 | 46.46 | 5.66 | 5.39 | 0.09 | 0.10 | 0.18 | 0.18 | 0.98 | 0.99 | 37.57 | 35.78 | 0.59 | 0.64 | 1.22 | 1.21 | 6.51 | 6.59 |
| 23.05.05 | 30.05.05 | 7 | 34.62 | 34.62 | 34.62 | 9.54 | 10.33 | 0.18 | 0.20 | 0.24 | 0.23 | 1.85 | 2.10 | 47.18 | 51.10 | 0.90 | 0.98 | 1.16 | 1.12 | 9.15 | 10.39 |
| 13.06.06 | 20.06.06 | 7 | 29.33 | 29.33 | 29.33 | 7.12 | 7.19 | 0.15 | 0.16 | 0.38 | 0.37 | 1.61 | 1.46 | 29.83 | 30.14 | 0.62 | 0.66 | 1.60 | 1.57 | 6.75 | 6.13 |
| 20.06.2006 | 27.06.2006 | 7 | 8.60 | 8.79 | 8.69 | 70.50 | 84.47 | 1.03 | 1.15 | 1.17 | 1.24 | 6.89 | 7.86 | 87.56 | 104.92 | 1.28 | 1.42 | 1.45 | 1.54 | 8.56 | 9.76 |

**Parallell Bergerhoff samples analused at EMD and LUA**

| | | | AVG 3 samples LUA | | | | EMD, one sample | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| From | To | days | As | Ni | Cd | Pb | As | Ni | Cd | Pb |
| 16-jan-06 | 13-feb-06 | 28 | 1.18 | 24.60 | 0.95 | 45.29 | 0.94 | 22.45 | 0.77 | 34.35 |
| 13-feb-06 | 13-mar-06 | 28 | 1.13 | 17.45 | 0.66 | 52.15 | 0.82 | 13.66 | 0.61 | 42.06 |
| 13-mar-06 | 10-apr-06 | 28 | 1.07 | 14.56 | 0.61 | 44.38 | 0.89 | 13.77 | 0.48 | 34.19 |
| 10-apr-06 | 9-mai-06 | 29 | 1.44 | 16.08 | 0.69 | 41.98 | 1.99 | 15.70 | 0.62 | 42.33 |
| 9-mai-06 | 6-jun-06 | 28 | 2.07 | 18.54 | 0.98 | 63.42 | 1.75 | 15.79 | 0.97 | 62.28 |
| 6-jun-06 | 4-jul-06 | 28 | 1.91 | 19.45 | 0.94 | 62.93 | 1.83 | 19.81 | 1.32 | 63.50 |

# A.4 Results from split samples from the field trials in Birkenes

**Split bulk samples analysed at NILU and UBA**

| | | | | | | concentrations, µg/l | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | Pb | Pb | Cd | Cd | As | As | Ni | Ni | Pb | Pb | Cd | Cd | As | As | Ni | Ni |
| **first day** | **last day** | **days** | collecto | **mm** | mm avg | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU |
| 20.11.06 | 27.11.06 | 7 | E | 142 | 145 | 0.48 | 0.49 | 0.01 | 0.01 | 0.05 | 0.06 | 0.18 | 0.14 | 9.69 | 9.98 | 0.26 | 0.22 | 1.10 | 1.16 | 3.57 | 2.95 |
| 20.11.06 | 27.11.06 | 7 | F | 148 | 145 | 0.37 | 0.48 | 0.01 | 0.01 | 0.05 | 0.04 | 0.13 | 0.12 | 7.83 | 10.16 | 0.23 | 0.23 | 1.13 | 0.89 | 2.70 | 2.45 |
| 25.12.06 | 01.01.07 | 7 | E | 75 | 75 | 1.01 | 1.02 | 0.02 | 0.02 | 0.08 | 0.08 | 0.22 | 0.16 | 10.81 | 10.91 | 0.26 | 0.23 | 0.84 | 0.87 | 2.37 | 1.73 |
| 25.12.06 | 01.01.07 | 7 | F | 75 | 75 | 0.97 | 1.07 | 0.03 | 0.02 | 0.08 | 0.08 | 0.18 | 0.16 | 10.31 | 11.37 | 0.27 | 0.26 | 0.86 | 0.86 | 1.88 | 1.76 |
| 01.01.07 | 08.01.07 | 7 | E | 28 | 27 | 0.78 | 0.88 | 0.01 | 0.01 | 0.08 | 0.07 | 0.06 | 0.18 | 3.14 | 3.55 | 0.04 | 0.04 | 0.31 | 0.29 | 0.25 | 0.74 |
| 01.01.07 | 08.01.07 | 7 | F | 26 | 27 | 0.83 | 0.89 | 0.01 | 0.01 | 0.08 | 0.06 | 0.25 | 0.20 | 3.07 | 3.28 | 0.05 | 0.04 | 0.28 | 0.23 | 0.94 | 0.74 |
| 05.02.07 | 12.02.07 | 7 | E | 16 | 16 | 0.64 | 0.69 | 0.02 | 0.02 | 0.10 | 0.09 | 0.41 | 0.33 | 1.45 | 1.57 | 0.05 | 0.05 | 0.22 | 0.20 | 0.93 | 0.74 |
| 12.03.07 | 19.03.07 | 7 | E | 18 | 18 | 0.22 | 0.20 | 0.00 | 0.00 | 0.06 | 0.03 | 0.35 | 0.27 | 0.56 | 0.52 | 0.01 | 0.01 | 0.16 | 0.08 | 0.89 | 0.68 |
| 16.04.07 | 23.04.07 | 7 | E | 11 | 11 | 0.99 | 1.08 | 0.04 | 0.03 | 0.21 | 0.24 | 2.22 | 1.78 | 1.53 | 1.66 | 0.06 | 0.05 | 0.32 | 0.37 | 3.43 | 2.75 |

**Split wet only samples analysed at NILU and UBA**

| | | | | | | concentrations, µg/l | | | | | | | | deposition µg/m2/day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | Pb | Pb | Cd | Cd | As | As | Ni | Ni | Pb | Pb | Cd | Cd | As | As | Ni | Ni |
| **first day** | **last day** | **days** | collecto | **mm** | mm avg | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA | NILU | UBA |
| 13.11.2006 | 20.11.2006 | 7 | G | 59 | 60 | 1.72 | 1.71 | 0.04 | 0.04 | 0.11 | 0.10 | 0.36 | 0.51 | 14.51 | 14.45 | 0.31 | 0.31 | 0.89 | 0.87 | 3.08 | 4.27 |
| 13.11.2006 | 20.11.2006 | 7 | H | 60 | 60 | 1.90 | 1.83 | 0.04 | 0.04 | 0.14 | 0.14 | 0.33 | 0.41 | 16.25 | 15.68 | 0.37 | 0.38 | 1.19 | 1.20 | 2.85 | 3.55 |
| 11.12.2006 | 18.12.2006 | 7 | G | 29 | 28 | 0.16 | 0.17 | 0.00 | 0.00 | 0.03 | 0.04 | 0.14 | 0.18 | 0.65 | 0.71 | 0.01 | 0.01 | 0.11 | 0.15 | 0.56 | 0.74 |
| 11.12.2006 | 18.12.2006 | 7 | H | 27 | 28 | 0.20 | 0.22 | 0.00 | 0.00 | 0.02 | 0.04 | 0.20 | 0.25 | 0.78 | 0.85 | 0.01 | 0.02 | 0.09 | 0.15 | 0.78 | 0.99 |
| 01.01.2007 | 08.01.2007 | 7 | G | 29 | 29 | 0.75 | 0.73 | 0.01 | 0.01 | 0.06 | 0.08 | 0.22 | 0.34 | 3.12 | 3.01 | 0.04 | 0.05 | 0.24 | 0.32 | 0.90 | 1.41 |
| 05.02.2007 | 12.02.2007 | 7 | G | 14 | 14 | 0.55 | 0.52 | 0.02 | 0.02 | 0.10 | 0.09 | 0.13 | 0.20 | 1.14 | 1.08 | 0.04 | 0.04 | 0.21 | 0.18 | 0.27 | 0.40 |
| 12.03.2007 | 19.03.2007 | 7 | G | 19 | 19 | 0.28 | 0.23 | 0.00 | 0.01 | 0.03 | 0.05 | 0.17 | 0.21 | 0.74 | 0.62 | 0.01 | 0.01 | 0.09 | 0.13 | 0.45 | 0.57 |
| 16.04.2007 | 23.04.2007 | 7 | G | 11 | 11 | 0.65 | 0.61 | 0.02 | 0.03 | 0.07 | 0.09 | 0.47 | 0.47 | 1.04 | 0.98 | 0.03 | 0.04 | 0.12 | 0.14 | 0.76 | 0.74 |

**Parallell Bergerhoff samples analysed at EMD and LUA**

| | | | AVG 3 samples EMD | | | | LUA, one sample | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| From | To | **days** | As | Ni | Cd | Pb | As | Ni | Cd | Pb |
| 1-nov-06 | 1-des-06 | 30 | 0.61 | 2.43 | 0.24 | 5.75 | 0.65 | 1.76 | 0.13 | 5.62 |
| 1-des-06 | 1-jan-07 | 31 | 0.39 | 1.21 | 0.11 | 1.69 | 0.34 | 1.70 | 0.07 | 1.76 |
| 1-jan-07 | 1-feb-07 | 31 | 0.53 | 1.16 | 0.10 | 1.28 | 0.18 | 1.57 | 0.01 | 1.79 |
| 1-feb-07 | 1-mar-07 | 28 | 0.14 | 0.46 | 0.07 | 1.37 | 0.11 | 0.72 | 0.03 | 1.11 |
| 1-mar-07 | 1-apr-07 | 31 | 0.20 | 0.84 | 0.09 | 1.74 | | | | |
| 1-apr-07 | 1-mai-07 | 30 | 0.09 | 0.52 | 0.03 | 0.80 | | | | |

# A.5 The bulk measurements at Peyrusse

| | | | mm | | Concentration Cd | | Pb | | As | | Ni | | | deposition, µg/m²day Cd | | Pb | | As | | Ni | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| first day | last day | nr of days | E | F | E | F | E | F | E | F | E | F | | E | F | E | F | E | F | E | F |
| 7-mar-06 | 14-mar-06 | 7 | 72.61 | 65.92 | 0.008 | 0.003 | 0.240 | 0.269 | 0.005 | 0.005 | 0.186 | 0.188 | | 0.086 | 0.024 | 2.492 | 2.533 | 0.052 | 0.047 | 1.933 | 1.773 |
| 14-mar-06 | 21-mar-06 | 7 | 2.87 | 3.03 | 0.047 | 0.036 | 2.935 | 3.548 | 0.343 | 0.177 | 1.299 | 1.873 | | 0.019 | 0.016 | 1.203 | 1.536 | 0.141 | 0.077 | 0.533 | 0.811 |
| 21-mar-06 | 28-mar-06 | 7 | 57.48 | 56.05 | 0.033 | 0.028 | 1.149 | 1.248 | 0.058 | 0.060 | 0.356 | 0.412 | | 0.270 | 0.228 | 9.436 | 9.993 | 0.475 | 0.482 | 2.927 | 3.303 |
| 28-mar-06 | 4-apr-06 | 7 | 3.98 | 3.82 | 0.161 | 0.054 | 1.569 | 1.384 | 0.126 | 0.099 | 0.555 | 0.501 | | 0.091 | 0.029 | 0.892 | 0.755 | 0.072 | 0.054 | 0.316 | 0.273 |
| rinsing | | | 6.37 | 6.37 | 0.023 | 0.058 | 2.413 | 3.777 | 0.005 | 0.005 | 0.452 | 2.602 | funnel | 0.005 | 0.013 | 0.549 | 0.859 | 0.001 | 0.001 | 0.103 | 0.592 |
| **07/03/2006-04/04/2006** | | | | | | | | | | | | | collector | 0.117 | 0.074 | 3.506 | 3.704 | 0.185 | 0.165 | 1.427 | 1.540 |
| | | | | | | | | | | | | | funnel + collector | 0.122 | 0.087 | 4.055 | 4.564 | 0.186 | 0.166 | 1.530 | 2.132 |
| 4-apr-06 | 11-apr-06 | 7 | 17.90 | 13.89 | 0.047 | 0.032 | 0.921 | 0.928 | 0.124 | 0.113 | 0.623 | 0.646 | | 0.120 | 0.064 | 2.355 | 1.841 | 0.316 | 0.225 | 1.594 | 1.281 |
| 11-apr-06 | 18-apr-06 | 7 | 2.87 | 3.15 | 0.122 | 0.033 | 2.470 | 1.800 | 0.217 | 0.128 | 1.190 | 0.643 | | 0.050 | 0.015 | 1.013 | 0.810 | 0.089 | 0.057 | 0.488 | 0.289 |
| 18-apr-06 | 24-apr-06 | 6 | 0.76 | 0.89 | 0.151 | 0.134 | 5.284 | 5.255 | 0.748 | 0.692 | 3.662 | 3.662 | | 0.019 | 0.020 | 0.669 | 0.780 | 0.095 | 0.103 | 0.464 | 0.543 |
| 24-apr-06 | 2-mai-06 | 8 | 10.61 | 10.51 | 0.030 | 0.025 | 0.806 | 0.955 | 0.195 | 0.141 | 0.587 | 0.510 | | 0.040 | 0.033 | 1.068 | 1.254 | 0.258 | 0.185 | 0.778 | 0.670 |
| rinsing | | | 6.37 | 6.37 | 0.045 | 0.108 | 2.531 | 2.354 | 0.140 | 0.130 | 0.928 | 0.557 | funnel | 0.010 | 0.025 | 0.576 | 0.536 | 0.032 | 0.029 | 0.211 | 0.127 |
| **04/04/2006-02/05/2006** | | | | | | | | | | | | | collector | 0.057 | 0.033 | 1.276 | 1.171 | 0.189 | 0.143 | 0.831 | 0.696 |
| | | | | | | | | | | | | | funnel + collector | 0.067 | 0.057 | 1.852 | 1.707 | 0.221 | 0.172 | 1.042 | 0.823 |
| 2-mai-06 | 9-mai-06 | 7 | 25.06 | 25.29 | 0.022 | 0.034 | 1.272 | 0.738 | 0.143 | 0.182 | 0.379 | 0.497 | | 0.079 | 0.124 | 4.552 | 2.668 | 0.512 | 0.659 | 1.358 | 1.797 |
| 9-mai-06 | 16-mai-06 | 7 | 15.38 | 15.99 | 0.039 | 0.035 | 1.296 | 1.710 | 0.194 | 0.184 | 0.560 | 0.506 | | 0.085 | 0.081 | 2.848 | 3.906 | 0.426 | 0.420 | 1.230 | 1.156 |
| 16-mai-06 | 23-mai-06 | 7 | 1.66 | 1.27 | 0.080 | 0.094 | 4.127 | 3.193 | 0.489 | 0.486 | 1.606 | 1.688 | | 0.019 | 0.017 | 0.979 | 0.579 | 0.116 | 0.088 | 0.381 | 0.306 |
| 23-mai-06 | 30-mai-06 | 7 | 0.54 | 0.54 | 0.089 | 0.079 | 1.936 | 1.995 | 0.571 | 0.311 | 1.671 | 1.415 | | 0.007 | 0.006 | 0.149 | 0.154 | 0.044 | 0.024 | 0.129 | 0.109 |
| rinsing | | | 6.37 | 6.37 | 0.038 | 0.087 | 2.633 | 3.165 | 0.147 | 0.115 | 1.398 | 0.545 | funnel | 0.009 | 0.020 | 0.599 | 0.720 | 0.034 | 0.026 | 0.318 | 0.124 |
| **02/05/2006-30/05/2006** | | | | | | | | | | | | | collector | 0.047 | 0.057 | 2.132 | 1.827 | 0.275 | 0.298 | 0.774 | 0.842 |
| | | | | | | | | | | | | | funnel + collector | 0.056 | 0.077 | 2.731 | 2.547 | 0.308 | 0.324 | 1.092 | 0.966 |
| 30-mai-06 | 6-jun-06 | 7 | | | | | | | | | | | | | | | | | | | |
| 6-jun-06 | 13-jun-06 | 7 | 0.64 | 0.22 | | | | | | | | | | | | | | | | | |
| 13-jun-06 | 20-jun-06 | 7 | 38.38 | 38.41 | 0.025 | 0.024 | 1.544 | 1.348 | 0.142 | 0.133 | 0.585 | 0.558 | | 0.138 | 0.134 | 8.464 | 7.395 | 0.776 | 0.733 | 3.208 | 3.062 |
| 20-jun-06 | 28-jun-06 | 8 | 15.86 | 8.31 | 0.048 | 0.150 | 1.570 | 1.834 | 0.214 | 0.252 | 1.182 | 1.741 | | 0.095 | 0.297 | 3.113 | 3.637 | 0.423 | 0.500 | 2.344 | 3.451 |
| rinsing | | | 6.37 | 6.37 | | | | | | | | | | | | | | | | | |
| **30/05/2006-28/06/2006** | | | | | | | | | | | | | collector | 0.116 | 0.215 | 5.788 | 5.516 | 0.600 | 0.616 | 2.776 | 3.256 |
| 28-jun-06 | 4-jul-06 | 6 | 0.00 | 0.00 | | | | | | | | | | | | | | | | | |
| 4-jul-06 | 8-aug-06 | 35 | 32.26 | 33.54 | 0.044 | 0.039 | 1.665 | 1.910 | 0.165 | 0.157 | 0.990 | 1.180 | | 0.041 | 0.037 | 1.535 | 1.830 | 0.152 | 0.150 | 0.913 | 1.131 |
| 8-aug-06 | 16-aug-06 | 8 | 1.40 | 4.65 | 0.057 | 0.044 | 2.943 | 5.441 | 0.300 | 0.406 | 2.516 | 6.119 | | 0.033 | 0.008 | 1.710 | 0.952 | 0.174 | 0.071 | 1.462 | 1.071 |
| 16-aug-06 | 22-aug-06 | 6 | 17.99 | 22.23 | 0.015 | 0.014 | 0.622 | 0.763 | 0.097 | 0.067 | 1.571 | 1.006 | | 0.045 | 0.051 | 1.864 | 2.828 | 0.291 | 0.249 | 4.711 | 3.726 |
| rinsing | | | 6.37 | 6.37 | | | | | | | | | | | | | | | | | |
| **28/06/2006-22/08/2006** | | | | | | | | | | | | | collector | 0.040 | 0.032 | 1.703 | 1.870 | 0.206 | 0.157 | 2.362 | 1.976 |

# A.6 The wet only measurements at Peyrusse

| | | | mm | | Concentration Cd | | Pb | | As | | Ni | | | deposition Cd | | Pb | | As | | Ni | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| first day | last day | days | G | H | G | H | G | H | G | H | G | H | | G | H | G | H | G | H | G | H | |
| 7-mar-06 | 14-mar-06 | 7 | 59.90 | 60.00 | 0.007 | 0.006 | 0.19 | 0.19 | 0.020 | 0.020 | 0.08 | 0.09 | | 0.068 | 0.063 | 1.892 | 1.903 | 0.195 | 0.195 | 0.790 | 0.861 | Overflow |
| 14-mar-06 | 21-mar-06 | 7 | 2.60 | 2.30 | 0.020 | 0.027 | 0.39 | 0.15 | 0.134 | 0.121 | 0.51 | 0.35 | | 0.008 | 0.009 | 0.144 | 0.049 | 0.050 | 0.040 | 0.190 | 0.115 | |
| 21-mar-06 | 28-mar-06 | 7 | 50.08 | 43.00 | 0.023 | 0.022 | 1.17 | 1.20 | 0.078 | 0.083 | 0.26 | 0.24 | | 0.162 | 0.133 | 8.351 | 7.395 | 0.557 | 0.507 | 1.854 | 1.490 | |
| 28-mar-06 | 4-apr-06 | 7 | 4.40 | 3.50 | 0.036 | 0.036 | 1.42 | 1.45 | 0.086 | 0.068 | 0.40 | 0.27 | | 0.022 | 0.018 | 0.894 | 0.726 | 0.054 | 0.034 | 0.249 | 0.135 | |
| rinsing | | | 4.00 | 4.00 | 0.002 | 0.010 | 0.86 | 1.63 | 0.008 | 0.014 | 0.05 | 0.07 | funnel | 0.000 | 0.001 | 0.122 | 0.233 | 0.001 | 0.002 | 0.008 | 0.011 | |
| **07/03/2006-04/04/2006** | | | | | | | | | | | | | wet dep | 0.065 | 0.056 | 2.820 | 2.518 | 0.214 | 0.194 | 0.771 | 0.650 | |
| | | | | | | | | | | | | | collector+ fu | 0.065 | 0.057 | 2.942 | 2.751 | 0.215 | 0.196 | 0.778 | 0.661 | |
| 4-apr-06 | 11-apr-06 | 7 | 15.50 | 14.20 | 0.029 | 0.031 | 1.03 | 1.13 | 0.102 | 0.117 | 0.31 | 0.36 | | 0.064 | 0.064 | 2.281 | 2.293 | 0.226 | 0.238 | 0.689 | 0.732 | |
| 11-apr-06 | 18-apr-06 | 7 | 3.60 | 2.80 | 0.043 | 0.064 | 1.63 | 1.96 | 0.109 | 0.130 | 0.59 | 0.75 | | 0.022 | 0.026 | 0.838 | 0.782 | 0.056 | 0.052 | 0.301 | 0.300 | |
| 18-apr-06 | 24-apr-06 | 6 | | | | | | | | | | | | | | | | | | | | |
| 24-apr-06 | 2-mai-06 | 8 | 11.40 | 10.80 | 0.022 | 0.024 | 1.10 | 0.93 | 0.112 | 0.112 | 0.37 | 0.39 | | 0.031 | 0.032 | 1.564 | 1.261 | 0.160 | 0.151 | 0.527 | 0.533 | |
| rinsing | | | 4.00 | 4.00 | 0.015 | 0.020 | 0.89 | 1.75 | 0.009 | 0.030 | 0.08 | 0.16 | funnel | 0.002 | 0.003 | 0.127 | 0.249 | 0.001 | 0.004 | 0.012 | 0.023 | |
| **04/04/2006-02/05/2006** | | | | | | | | | | | | | wet dep | 0.039 | 0.041 | 1.561 | 1.445 | 0.147 | 0.147 | 0.506 | 0.522 | |
| | | | | | | | | | | | | | collector+ fu | 0.041 | 0.043 | 1.687 | 1.695 | 0.149 | 0.151 | 0.518 | 0.545 | |
| 2-mai-06 | 9-mai-06 | 7 | 27.20 | 25.00 | 0.021 | 0.023 | 0.93 | 1.35 | 0.072 | 0.081 | 0.22 | 0.24 | | 0.080 | 0.081 | 3.618 | 4.810 | 0.280 | 0.289 | 0.839 | 0.862 | |
| 9-mai-06 | 16-mai-06 | 7 | 18.50 | 15.90 | 0.043 | 0.048 | 1.83 | 2.21 | 0.128 | 0.173 | 0.53 | 0.56 | | 0.115 | 0.109 | 4.834 | 5.030 | 0.338 | 0.392 | 1.393 | 1.263 | |
| 16-mai-06 | 23-mai-06 | 7 | 2.60 | 2.60 | 0.055 | | 2.20 | | 0.330 | | 0.94 | | | 0.020 | 0.020 | 0.816 | 0.816 | 0.123 | 0.123 | 0.348 | 0.348 | |
| 23-mai-06 | 30-mai-06 | 7 | | | | | | | | | | | | | | | | | | | | |
| rinsing | | | 4.00 | 4.00 | 0.009 | 0.015 | 0.65 | 1.52 | 0.014 | 0.029 | 0.11 | 0.14 | funnel | 0.001 | 0.002 | 0.092 | 0.217 | 0.002 | 0.004 | 0.016 | 0.020 | |
| **02/05/2006-30/05/2006** | | | | | | | | | | | | | wet dep | 0.072 | 0.070 | 3.089 | 3.552 | 0.247 | 0.268 | 0.860 | 0.824 | |
| | | | | | | | | | | | | | collector+ fu | 0.073 | 0.072 | 3.182 | 3.769 | 0.249 | 0.272 | 0.876 | 0.844 | |
| 30-mai-06 | 6-jun-06 | 7 | | | | | | | | | | | | | | | | | | | | |
| 6-jun-06 | 13-jun-06 | 7 | | | | | | | | | | | | | | | | | | | | |
| 13-jun-06 | 20-jun-06 | 7 | 42.90 | 40.00 | 0.031 | 0.032 | 1.12 | 1.25 | 0.097 | 0.118 | 0.42 | 0.47 | | 0.192 | 0.182 | 6.872 | 7.154 | 0.591 | 0.673 | 2.603 | 2.659 | |
| 20-jun-06 | 28-jun-06 | 8 | 20.00 | 15.90 | 0.040 | 0.049 | 1.80 | 1.83 | 0.151 | 0.176 | 0.52 | 0.62 | | 0.101 | 0.097 | 4.490 | 3.645 | 0.377 | 0.349 | 1.312 | 1.241 | |
| rinsing | | | 4.00 | 4.00 | 0.005 | 0.015 | 0.93 | 1.96 | 0.013 | 0.026 | 0.09 | 0.18 | funnel | 0.001 | 0.002 | 0.129 | 0.270 | 0.002 | 0.004 | 0.012 | 0.024 | |
| **30/05/2006-28/06/2006** | | | | | | | | | | | | | wet dep | 0.146 | 0.140 | 5.681 | 5.399 | 0.484 | 0.511 | 1.958 | 1.950 | |
| | | | | | | | | | | | | | collector+ fu | 0.147 | 0.142 | 5.810 | 5.669 | 0.486 | 0.514 | 1.970 | 1.974 | |
| 28.06.06 | 04.07.06 | 6 | | | | | | | | | | | | | | | | | | | | |
| 04.07.06 | 11.07.06 | 7 | 5.2 | 5.0 | 0.025 | 0.034 | 1.36 | 1.86 | 0.120 | 0.142 | 0.77 | 1.28 | | 0.018 | 0.024 | 1.010 | 1.328 | 0.089 | 0.102 | 0.574 | 0.912 | |
| 11.07.06 | 18.07.06 | 7 | 2.9 | 2.3 | 0.039 | 0.035 | 2.59 | 1.91 | 0.237 | 0.196 | 1.24 | 1.17 | | 0.016 | 0.011 | 1.075 | 0.628 | 0.098 | 0.064 | 0.515 | 0.385 | |
| 18.07.06 | 25.07.06 | 7 | 16.8 | 15.7 | 0.022 | 0.021 | 1.23 | 1.19 | 0.082 | 0.089 | 0.42 | 0.45 | | 0.053 | 0.047 | 2.943 | 2.673 | 0.196 | 0.200 | 1.010 | 1.001 | |
| rinsing | | | 4.00 | 4.00 | 0.002 | 0.005 | 0.48 | 0.41 | 0.004 | 0.008 | 0.08 | 0.07 | funnel | 0.000 | 0.001 | 0.071 | 0.061 | 0.001 | 0.001 | 0.012 | 0.011 | |
| **28/06/2006-25/07/2006** | | | | | | | | | | | | | wet dep | 0.029 | 0.027 | 1.676 | 1.543 | 0.128 | 0.122 | 0.700 | 0.766 | |
| | | | | | | | | | | | | | collector+ fu | 0.029 | 0.028 | 1.747 | 1.604 | 0.128 | 0.123 | 0.712 | 0.777 | |
| 25.07.06 | 01.08.06 | 7 | 8.6 | 8.4 | 0.010 | 0.011 | 0.81 | 0.60 | 0.056 | 0.059 | 1.11 | 0.92 | | 0.012 | 0.013 | 0.991 | 0.722 | 0.069 | 0.071 | 1.366 | 1.110 | |
| 01.08.06 | 08.08.06 | 7 | 5.1 | 4.6 | 0.006 | 0.006 | 0.68 | 0.48 | 0.026 | 0.023 | 0.28 | 0.17 | | 0.004 | 0.004 | 0.495 | 0.313 | 0.019 | 0.015 | 0.207 | 0.109 | |
| 08.08.06 | 16.08.06 | 8 | 5.2 | 4.9 | 0.017 | 0.022 | 0.92 | 0.94 | 0.149 | 0.163 | 0.51 | 0.77 | | 0.011 | 0.013 | 0.598 | 0.575 | 0.097 | 0.100 | 0.334 | 0.471 | |
| 16.08.06 | 22.08.06 | 6 | 21.0 | 20.5 | 0.012 | 0.008 | 0.46 | 0.40 | 0.056 | 0.053 | 0.19 | 0.21 | | 0.042 | 0.027 | 1.619 | 1.353 | 0.196 | 0.180 | 0.674 | 0.703 | |
| rinsing | | | 4.00 | 4.00 | 0.002 | 0.001 | 0.35 | 0.38 | 0.006 | 0.004 | 0.21 | 0.13 | funnel | 0.000 | 0.000 | 0.050 | 0.054 | 0.001 | 0.001 | 0.030 | 0.018 | |
| **25/07/2006-22/08/2006** | | | | | | | | | | | | | wet dep | 0.017 | 0.014 | 0.926 | 0.741 | 0.095 | 0.091 | 0.645 | 0.598 | |
| | | | | | | | | | | | | | collector+ fu | 0.018 | 0.015 | 0.976 | 0.795 | 0.096 | 0.092 | 0.676 | 0.616 | |

## A.7 The Bergerhoff measurements at Peyrusse

| | | | Deposition flux in µg/m²/d | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | As | | | Ni | | | Cd | | | Pb | | |
| From | To | nr of days | A | C | D | A | C | D | A | C | D | A | C | D |
| 07.03.2006 | 04.04.2006 | 28 | 0.213 | 0.240 | 0.279 | 0.668 | 0.706 | 0.732 | 0.057 | 0.074 | 0.059 | 2.004 | 2.387 | 2.527 |
| 04.04.2006 | 02.05.2006 | 28 | 0.233 | 0.276 | 0.249 | 0.876 | 0.963 | 0.975 | 0.070 | 0.053 | 0.053 | 2.046 | 1.872 | 1.948 |
| 02.05.2006 | 30.05.2006 | 28 | 0.246 | 0.232 | 0.227 | 0.828 | 0.845 | 0.910 | 0.060 | 0.052 | 0.058 | 2.600 | 1.839 | 2.336 |
| 30.05.2006 | 28.06.2006 | 29 | 0.652 | 0.725 | 0.730 | 2.354 | 2.566 | 2.755 | 0.082 | 0.085 | 0.093 | 3.567 | 3.778 | 4.237 |
| 28.06.2006 | 25.07.2006 | 27 | 0.161 | 0.143 | 0.178 | 0.554 | 0.583 | 0.590 | 0.032 | 0.034 | 0.040 | 1.564 | 1.793 | 1.769 |
| 25.07.2006 | 22.08.2006 | 28 | 0.187 | 0.201 | 0.174 | 0.433 | 0.466 | 0.480 | 0.020 | 0.018 | 0.016 | 1.636 | 1.423 | 1.553 |

## A.8 The bulk measurements at Copenhagen

| | | | | | Concentration, µg/l | | | | | | | | | deposition, µg/m²day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | mm | | Cd | | Pb | | As | | Ni | | | Cd | | Pb | | As | | Ni | |
| first day | last day | nr of days | A | B | A | B | A | B | A | B | A | B | | A | B | A | B | A | B | A | B |
| 12-sep-06 | 19-sep-06 | 7 | 0.04 | 0.04 | 0.392 | 0.437 | 20.18 | 23.80 | 1.486 | 1.658 | 6.19 | 6.08 | | 0.002 | 0.003 | 0.125 | 0.147 | 0.009 | 0.010 | 0.038 | 0.038 |
| 19-sep-06 | 26-sep-06 | 7 | 0.01 | 0.00 | | | | | | | | | | | | | | | | | |
| 26-sep-06 | 3-okt-06 | 7 | 22.42 | 21.92 | 0.039 | 0.044 | 2.03 | 1.89 | 0.172 | 0.173 | 0.40 | 0.40 | | 0.124 | 0.139 | 6.508 | 6.059 | 0.550 | 0.554 | 1.267 | 1.267 |
| rinsing | | | 6.37 | 6.37 | 0.038 | 0.050 | 5.64 | 5.75 | 0.170 | 0.189 | 0.58 | 0.67 | funnel | 0.011 | 0.015 | 1.711 | 1.744 | 0.052 | 0.057 | 0.176 | 0.202 |
| **05/09/2006-03/10/2006** | | | | | | | | | | | | | collector | 0.063 | 0.071 | 3.316 | 3.103 | 0.280 | 0.282 | 0.652 | 0.652 |
| | | | | | | | | | | | | | funnel + collector | 0.075 | 0.086 | 5.028 | 4.847 | 0.331 | 0.340 | 0.828 | 0.854 |
| 3-okt-06 | 10-okt-06 | 7 | 14.04 | 14.59 | 0.033 | 0.029 | 1.21 | 0.88 | 0.060 | 0.056 | 0.24 | 0.19 | | 0.066 | 0.059 | 2.425 | 1.759 | 0.120 | 0.113 | 0.485 | 0.388 |
| 10-okt-06 | 17-okt-06 | 7 | 0.08 | 0.04 | 0.038 | 0.205 | 11.81 | 11.23 | 0.140 | 0.170 | 7.10 | 5.11 | | 0.000 | 0.002 | 0.134 | 0.128 | 0.002 | 0.002 | 0.081 | 0.058 |
| 17-okt-06 | 24-okt-06 | 7 | 15.69 | 16.02 | 0.046 | 0.026 | 1.30 | 1.01 | 0.061 | 0.066 | 0.290 | 0.241 | | 0.104 | 0.057 | 2.906 | 2.254 | 0.136 | 0.147 | 0.650 | 0.540 |
| 24-okt-06 | 1-nov-06 | 8 | 34.96 | 35.64 | 0.032 | 0.037 | 1.50 | 1.50 | 0.106 | 0.118 | 0.331 | 0.450 | | 0.139 | 0.160 | 6.534 | 6.561 | 0.462 | 0.516 | 1.446 | 1.965 |
| rinsing | | | 6.37 | 6.37 | 0.052 | 0.023 | 4.34 | 3.33 | 0.051 | 0.051 | 0.412 | 0.310 | funnel | 0.011 | 0.005 | 0.953 | 0.731 | 0.011 | 0.011 | 0.090 | 0.068 |
| **03/10/2006-01/11/2006** | | | | | | | | | | | | | collector | 0.077 | 0.070 | 3.000 | 2.676 | 0.180 | 0.195 | 0.665 | 0.738 |
| | | | | | | | | | | | | | funnel + collector | 0.088 | 0.075 | 3.953 | 3.406 | 0.191 | 0.206 | 0.756 | 0.806 |
| 1-nov-06 | 7-nov-06 | 6 | 8.36 | 8.34 | 0.021 | | 0.98 | | 0.130 | | 0.474 | | | 0.030 | 0.030 | 1.365 | 1.365 | 0.181 | 0.181 | 0.660 | 0.660 |
| 7-nov-06 | 15-nov-06 | 8 | 33.05 | 33.25 | 0.021 | | 1.21 | | 0.074 | | 0.202 | | | 0.088 | 0.088 | 5.014 | 5.014 | 0.305 | 0.305 | 0.833 | 0.833 |
| 15-nov-06 | 21-nov-06 | 6 | 16.09 | 16.47 | 0.033 | 0.032 | 1.58 | 1.87 | 0.141 | 0.160 | 1.389 | 1.304 | | 0.088 | 0.087 | 4.238 | 5.010 | 0.378 | 0.430 | 3.725 | 3.496 |
| 21-nov-06 | 28-nov-06 | 7 | 11.64 | 11.79 | 0.028 | 0.028 | 1.33 | 1.01 | 0.071 | 0.081 | 1.301 | 1.293 | | 0.046 | 0.046 | 2.215 | 1.673 | 0.117 | 0.135 | 2.163 | 2.151 |
| rinsing | | | 6.37 | 6.37 | 0.020 | 0.065 | 3.23 | 3.20 | 0.038 | 0.033 | 0.457 | 0.407 | funnel | 0.005 | 0.015 | 0.762 | 0.755 | 0.009 | 0.008 | 0.108 | 0.096 |
| **01/11/2006-28/11/2006** | | | | | | | | | | | | | collector | 0.063 | 0.063 | 3.208 | 3.265 | 0.245 | 0.263 | 1.845 | 1.785 |
| | | | | | | | | | | | | | funnel + collector | 0.068 | 0.078 | 3.970 | 4.020 | 0.254 | 0.270 | 1.953 | 1.881 |
| 28-nov-06 | 5-des-06 | 7 | 9.63 | 9.64 | 0.044 | 0.042 | 1.79 | 1.79 | 0.122 | 0.111 | 0.385 | 0.537 | | 0.060 | 0.058 | 2.464 | 2.469 | 0.168 | 0.152 | 0.529 | 0.739 |
| 5-des-06 | 12-des-06 | 7 | 13.63 | 13.42 | 0.045 | 0.049 | 1.68 | 1.67 | 0.060 | 0.044 | 0.238 | 0.228 | | 0.087 | 0.095 | 3.276 | 3.246 | 0.117 | 0.085 | 0.463 | 0.445 |
| 12-des-06 | 19-des-06 | 7 | 25.86 | 25.66 | 0.031 | 0.029 | 1.14 | 1.20 | 0.086 | 0.094 | 0.461 | 0.447 | | 0.114 | 0.106 | 4.223 | 4.435 | 0.316 | 0.346 | 1.703 | 1.650 |
| 19-des-06 | 27-des-06 | 8 | 0.11 | 0.09 | 0.205 | 0.243 | 4.84 | 8.94 | 0.724 | 0.664 | 7.88 | 10.84 | | 0.003 | 0.003 | 0.064 | 0.117 | 0.010 | 0.009 | 0.103 | 0.142 |
| rinsing | | | 6.37 | 6.37 | 0.033 | 0.02 | 2.214 | 1.936 | 0.031 | 0.023 | 0.295 | 0.373 | funnel | 0.007 | 0.004 | 0.486 | 0.425 | 0.007 | 0.005 | 0.065 | 0.082 |
| **28/11/2006-27/12/2006** | | | | | | | | | | | | | collector | 0.066 | 0.065 | 2.507 | 2.567 | 0.152 | 0.148 | 0.700 | 0.744 |
| | | | | | | | | | | | | | funnel + collector | 0.073 | 0.070 | 2.993 | 2.992 | 0.159 | 0.153 | 0.765 | 0.826 |
| 27-des-06 | 2-jan-07 | 6 | 24.18 | 24.39 | 0.020 | 0.034 | 1.68 | 1.36 | 0.046 | 0.036 | 0.130 | 0.137 | | 0.081 | 0.135 | 6.758 | 5.466 | 0.185 | 0.146 | 0.523 | 0.554 |
| 2-jan-07 | 9-jan-07 | 7 | 14.45 | 14.64 | 0.042 | 0.035 | 3.82 | 3.26 | 0.139 | 0.112 | 0.574 | 0.485 | | 0.086 | 0.072 | 7.893 | 6.726 | 0.286 | 0.231 | 1.185 | 1.001 |
| 9-jan-07 | 16-jan-07 | 7 | 17.44 | 18.02 | 0.026 | 0.022 | 0.87 | 0.98 | 0.144 | 0.143 | 0.358 | 0.323 | | 0.068 | 0.056 | 2.228 | 2.451 | 0.372 | 0.356 | 0.922 | 0.805 |
| 16-jan-07 | 24-jan-07 | 8 | 42.97 | 43.72 | 0.010 | 0.011 | 0.43 | 0.41 | 0.049 | 0.042 | 0.213 | 0.234 | | 0.053 | 0.057 | 2.312 | 2.201 | 0.262 | 0.223 | 1.142 | 1.257 |
| rinsing | | | 6.37 | 6.37 | 0.014 | 0.050 | 3.98 | 4.19 | 0.036 | 0.043 | 0.255 | 0.188 | funnel | 0.003 | 0.011 | 0.906 | 0.954 | 0.008 | 0.010 | 0.058 | 0.043 |
| **27/12/2006-24/01/2007** | | | | | | | | | | | | | collector | 0.072 | 0.080 | 4.798 | 4.211 | 0.276 | 0.239 | 0.943 | 0.904 |
| | | | | | | | | | | | | | funnel + collector | 0.075 | 0.092 | 5.704 | 5.165 | 0.285 | 0.249 | 1.001 | 0.947 |
| 24-jan-07 | 30-jan-07 | 6 | 13.82 | 13.70 | 0.018 | 0.014 | 0.31 | 0.48 | 0.060 | 0.071 | 0.419 | 0.248 | | 0.042 | 0.032 | 0.709 | 1.106 | 0.138 | 0.163 | 0.964 | 0.570 |
| 30-jan-07 | 6-feb-07 | 7 | 10.26 | 10.29 | 0.020 | 0.018 | 0.66 | 0.83 | 0.117 | 0.123 | 0.533 | 0.414 | | 0.030 | 0.026 | 0.960 | 1.212 | 0.171 | 0.180 | 0.781 | 0.607 |
| 6-feb-07 | 13-feb-07 | 7 | 8.98 | 9.14 | 0.155 | 0.157 | 5.68 | 5.19 | 0.570 | 0.558 | 0.787 | 0.610 | | 0.199 | 0.201 | 7.291 | 6.662 | 0.731 | 0.716 | 1.010 | 0.783 |
| 13-feb-07 | 20-feb-07 | 7 | 1.09 | 1.16 | 0.219 | 0.237 | 8.00 | 7.11 | 0.515 | 0.470 | 0.787 | 0.610 | | 0.034 | 0.037 | 1.243 | 1.105 | 0.080 | 0.073 | 0.122 | 0.095 |
| rinsing | | | 6.37 | 6.37 | 0.021 | 0.022 | 2.48 | 3.53 | 0.046 | 0.071 | 0.359 | 0.845 | funnel | 0.005 | 0.005 | 0.586 | 0.833 | 0.011 | 0.017 | 0.085 | 0.199 |
| **24/01/2007-20/02/2007** | | | | | | | | | | | | | collector | 0.076 | 0.074 | 2.551 | 2.521 | 0.280 | 0.283 | 0.719 | 0.514 |
| | | | | | | | | | | | | | funnel + collector | 0.081 | 0.079 | 3.137 | 3.355 | 0.291 | 0.300 | 0.804 | 0.713 |

## A.9 The wet only measurements at Copenhagen

| | | | | | Concentration, µg/l | | | | | | | | | deposition | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | mm | | Cd | | Pb | | As | | Ni | | | Cd | | Pb | | As | | Ni | |
| **first day** | **last day** | nr of days | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 |
| 5-sep-06 | 12-sep-06 | 7 | 0.55 | 5.75 | | 0.041 | | 1.817 | | 0.134 | | 1.369 | | 0.034 | 0.034 | 1.491 | 1.491 | 0.110 | 0.110 | 1.124 | 1.124 |
| 12-sep-06 | 19-sep-06 | 7 | 0.30 | 0.19 | | | | | | | | | | | | | | | | | |
| 19-sep-06 | 26-sep-06 | 7 | 0.00 | 0.00 | | | | | | | | | | | | | | | | | |
| 26-sep-06 | 3-okt-06 | 7 | 26.07 | 24.86 | 0.030 | 0.030 | 1.196 | 1.256 | 0.097 | 0.106 | 0.358 | 0.397 | | 0.112 | 0.108 | 4.454 | 4.462 | 0.362 | 0.378 | 1.332 | 1.410 |
| rinsing | | | 4.00 | 4.00 | 0.006 | 0.008 | 0.754 | 0.872 | 0.024 | 0.024 | 0.138 | 0.151 | funnel | 0.001 | 0.001 | 0.108 | 0.125 | 0.003 | 0.003 | 0.020 | 0.022 |
| **05/09/2006-03/10/2006** | | | | | | | | | | | | | wet dep | 0.112 | 0.108 | 4.454 | 4.462 | 0.362 | 0.378 | 1.332 | 1.410 |
| | | | | | | | | | | | | | collector+ fu | 0.113 | 0.109 | 4.562 | 4.586 | 0.365 | 0.381 | 1.352 | 1.432 |
| 3-okt-06 | 10-okt-06 | 7 | 17.12 | 16.06 | 0.028 | 0.022 | 0.602 | 0.866 | 0.056 | 0.052 | 0.239 | 0.469 | | 0.069 | 0.050 | 1.473 | 1.986 | 0.136 | 0.120 | 0.583 | 1.075 |
| 10-okt-06 | 17-okt-06 | 7 | 0.00 | 0.00 | | | | | | | | | | | | | | | | | |
| 17-okt-06 | 24-okt-06 | 7 | 17.92 | 16.78 | 0.020 | 0.033 | 0.937 | 0.955 | 0.056 | 0.051 | 0.409 | 0.461 | | 0.052 | 0.080 | 2.398 | 2.289 | 0.142 | 0.121 | 1.048 | 1.105 |
| 24-okt-06 | 1-nov-06 | 8 | 38.47 | 36.62 | 0.029 | 0.031 | 1.330 | 1.673 | 0.106 | 0.110 | 0.335 | 0.371 | | 0.140 | 0.144 | 6.397 | 7.657 | 0.511 | 0.504 | 1.609 | 1.699 |
| rinsing | | | 4.00 | 4.00 | 0.004 | 0.003 | 1.065 | 0.795 | 0.010 | 0.012 | 0.103 | 0.193 | funnel | 0.000 | 0.000 | 0.147 | 0.110 | 0.001 | 0.002 | 0.014 | 0.027 |
| **03/10/2006-01/11/2006** | | | | | | | | | | | | | wet dep | 0.087 | 0.091 | 3.423 | 3.977 | 0.263 | 0.248 | 1.080 | 1.293 |
| | | | | | | | | | | | | | collector+ fu | 0.087 | 0.091 | 3.570 | 4.087 | 0.265 | 0.250 | 1.094 | 1.320 |
| 1-nov-06 | 7-nov-06 | 6 | 8.09 | 8.35 | 0.013 | 0.015 | 0.722 | 1.165 | 0.119 | 0.119 | 0.322 | 0.679 | | 0.018 | 0.021 | 0.973 | 1.621 | 0.161 | 0.165 | 0.433 | 0.944 |
| 7-nov-06 | 15-nov-06 | 8 | 33.66 | 34.09 | 0.024 | 0.023 | 1.537 | 1.574 | 0.095 | 0.093 | 0.311 | 0.373 | | 0.101 | 0.100 | 6.466 | 6.709 | 0.399 | 0.398 | 1.307 | 1.590 |
| 15-nov-06 | 21-nov-06 | 6 | 16.20 | 16.30 | 0.029 | 0.027 | 1.336 | 1.366 | 0.145 | 0.151 | 0.476 | 0.704 | | 0.078 | 0.072 | 3.608 | 3.712 | 0.390 | 0.409 | 1.285 | 1.912 |
| 21-nov-06 | 28-nov-06 | 7 | 11.25 | 11.13 | 0.032 | 0.037 | 1.068 | 1.076 | 0.080 | 0.086 | 1.232 | 1.425 | | 0.052 | 0.058 | 1.716 | 1.712 | 0.129 | 0.137 | 1.981 | 2.267 |
| rinsing | | | 4.00 | 4.00 | 0.006 | 0.005 | 1.466 | 1.410 | 0.013 | 0.017 | 0.112 | 0.173 | funnel | 0.001 | 0.001 | 0.217 | 0.209 | 0.002 | 0.002 | 0.017 | 0.026 |
| **01/11/2006-28/11/2006** | | | | | | | | | | | | | wet dep | 0.062 | 0.063 | 3.191 | 3.439 | 0.270 | 0.277 | 1.251 | 1.678 |
| | | | | | | | | | | | | | collector+ fu | 0.063 | 0.064 | 3.408 | 3.647 | 0.272 | 0.280 | 1.268 | 1.704 |
| 28-nov-06 | 5-des-06 | 7 | 9.74 | 9.41 | 0.031 | 0.033 | 0.830 | 1.016 | 0.094 | 0.095 | | 0.607 | | 0.044 | 0.044 | 1.155 | 1.364 | 0.131 | 0.127 | 0.815 | 0.815 |
| 5-des-06 | 12-des-06 | 7 | 13.20 | 12.93 | 0.048 | 0.047 | 1.955 | 1.718 | 0.109 | 0.114 | 0.544 | 0.623 | | 0.091 | 0.087 | 3.685 | 3.172 | 0.206 | 0.210 | 1.026 | 1.150 |
| 12-des-06 | 19-des-06 | 7 | 22.06 | 22.38 | 0.034 | 0.034 | 1.288 | 1.335 | 0.112 | 0.124 | 0.621 | 0.670 | | 0.108 | 0.109 | 4.060 | 4.266 | 0.353 | 0.398 | 1.958 | 2.142 |
| 19-des-06 | 27-des-06 | 8 | 0.00 | 0.00 | | | | | | | | | | | | | | | | | |
| rinsing | | | 4.00 | 4.00 | 0.006 | 0.005 | 1.144 | 1.366 | 0.019 | 0.020 | 0.518 | 0.177 | funnel | 0.001 | 0.001 | 0.158 | 0.188 | 0.003 | 0.003 | 0.071 | 0.024 |
| **28/11/2006-27/12/2006** | | | | | | | | | | | | | wet dep | 0.081 | 0.080 | 2.967 | 2.934 | 0.230 | 0.245 | 1.266 | 1.369 |
| | | | | | | | | | | | | | collector+ fu | 0.082 | 0.081 | 3.125 | 3.123 | 0.233 | 0.248 | 1.338 | 1.393 |
| 27-des-06 | 2-jan-07 | 6 | 24.57 | 24.20 | 0.022 | 0.025 | 1.287 | 1.302 | 0.056 | 0.046 | 0.432 | | | 0.092 | 0.101 | 5.268 | 5.253 | 0.228 | 0.184 | 1.767 | 1.767 |
| 2-jan-07 | 9-jan-07 | 7 | 12.88 | 12.58 | 0.039 | 0.036 | 1.671 | 1.205 | 0.114 | 0.125 | 0.818 | 0.727 | | 0.072 | 0.064 | 3.074 | 2.166 | 0.210 | 0.225 | 1.504 | 1.307 |
| 9-jan-07 | 16-jan-07 | 7 | 16.38 | 15.91 | 0.022 | 0.023 | 0.839 | 0.839 | 0.135 | 0.145 | 0.359 | 0.543 | | 0.052 | 0.052 | 1.964 | 1.908 | 0.315 | 0.328 | 0.839 | 1.233 |
| 16-jan-07 | 24-jan-07 | 8 | 43.24 | 43.51 | 0.009 | 0.009 | 0.430 | 0.430 | 0.049 | 0.048 | 0.227 | 0.353 | | 0.051 | 0.049 | 2.322 | 2.336 | 0.266 | 0.259 | 1.224 | 1.919 |
| rinsing | | | 4.00 | 4.00 | 0.007 | 0.006 | 1.891 | 1.599 | 0.017 | 0.023 | 0.142 | 0.212 | funnel | 0.001 | 0.001 | 0.270 | 0.228 | 0.002 | 0.003 | 0.020 | 0.030 |
| **27/12/2006-24/01/2007** | | | | | | | | | | | | | wet dep | 0.067 | 0.066 | 3.157 | 2.916 | 0.255 | 0.249 | 1.334 | 1.556 |
| | | | | | | | | | | | | | collector+ fu | 0.067 | 0.067 | 3.427 | 3.144 | 0.257 | 0.252 | 1.354 | 1.587 |
| 24-jan-07 | 30-jan-07 | 6 | 12.00 | 12.45 | 0.015 | 0.015 | 0.464 | 0.566 | 0.081 | 0.080 | | 0.355 | | 0.030 | 0.031 | 0.927 | 1.176 | 0.162 | 0.166 | 0.736 | 0.736 |
| 30-jan-07 | 6-feb-07 | 7 | 9.23 | 7.98 | 0.017 | 0.016 | 0.564 | 0.395 | 0.117 | 0.090 | 0.409 | 0.315 | | 0.022 | 0.018 | 0.743 | 0.451 | 0.154 | 0.103 | 0.539 | 0.359 |
| 6-feb-07 | 13-feb-07 | 7 | 8.28 | | 0.173 | | 6.737 | | 0.650 | | 0.884 | | | 0.205 | 0.205 | 7.973 | 7.973 | 0.769 | 0.769 | 1.046 | 1.046 |
| 13-feb-07 | 20-feb-07 | 7 | 0.52 | | | | | | | | | | | | | | | | | | |
| rinsing | | | 4.00 | 4.00 | 0.011 | 0.012 | 0.882 | 0.885 | 0.028 | 0.027 | 0.374 | 0.387 | funnel | 0.002 | 0.002 | 0.131 | 0.131 | 0.004 | 0.004 | 0.055 | 0.057 |
| 24/01/2007-20/02/2007 | | | | | | | | | | | | | wet dep | 0.086 | 0.084 | 3.215 | 3.200 | 0.362 | 0.346 | 0.773 | 0.713 |
| | | | | | | | | | | | | | collector+ fu | 0.087 | 0.086 | 3.345 | 3.331 | 0.366 | 0.350 | 0.829 | 0.771 |

## A.10 The bergerhoff measurements at Copenhagen

| | | | Deposition flux in µg/m²/d | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | **As** | | | **Ni** | | | **Cd** | | | **Pb** | | |
| From | To | nr of days | LUA | LUA | LUA | LUA | LUA | LUA | LUA | LUA | LUA | LUA | LUA | LUA |
| 05.09.2006 | 03.10.2006 | 28 | 0.485 | 0.235 | 0.542 | 1.334 | 0.973 | 0.965 | 0.110 | 0.060 | 0.142 | 5.676 | 4.384 | 42.364 |
| 03.10.2006 | 01.11.2006 | 29 | 0.445 | 0.482 | 0.473 | 2.159 | 1.861 | 1.614 | 0.079 | 0.131 | 0.127 | 5.841 | 33.467 | 5.883 |
| 01.11.2006 | 28.11.2006 | 27 | 0.384 | 0.743 | 0.350 | 3.464 | 2.782 | 3.628 | 0.084 | 0.062 | 0.105 | 11.363 | 3.474 | 14.097 |
| 28.11.2006 | 27.12.2006 | 29 | 0.117 | 0.267 | 0.369 | 1.989 | 1.751 | 1.869 | 0.129 | 0.096 | 0.187 | 6.647 | 7.668 | 4.706 |
| 27.12.2006 | 24.01.2007 | 28 | 0.316 | 0.249 | 0.191 | 2.273 | 2.230 | 3.550 | 0.087 | 0.127 | 0.114 | 11.362 | 10.945 | 8.363 |
| 24.01.2007 | 20.02.2007 | 27 | 0.363 | 0.473 | 0.179 | 5.128 | 2.905 | 2.445 | 0.117 | 0.101 | 0.143 | 6.273 | 5.510 | 6.968 |

# A.11 The bulk measurements at Duisburg

| | | | mm | | Concentration Cd | | Pb | | As | | Ni | | | deposition, µg/m²day Cd | | Pb | | As | | Ni | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **first day** | **last day** | Days | E | F | E | F | E | F | E | F | E | F | | E | F | E | F | E | F | E | F |
| 16-jan-06 | 23-jan-06 | 7 | 12.7 | 13.1 | 0.129 | 0.147 | 5.170 | 6.100 | 0.199 | 0.214 | 1.391 | 1.610 | | 0.233 | 0.275 | 9.352 | 11.422 | 0.360 | 0.400 | 2.515 | 3.015 |
| 23-jan-06 | 30-jan-06 | 7 | 3.2 | 3.3 | 0.390 | 0.363 | 17.234 | 16.788 | 0.453 | 0.435 | 6.600 | 5.495 | | 0.176 | 0.171 | 7.786 | 7.890 | 0.205 | 0.205 | 2.982 | 2.583 |
| 30-jan-06 | 6-feb-06 | 7 | 3.8 | 3.9 | 1.972 | 1.832 | 113.650 | 107.494 | 1.650 | 1.471 | 16.455 | 14.731 | | 1.073 | 1.012 | 61.841 | 59.420 | 0.898 | 0.813 | 8.954 | 8.143 |
| 6-feb-06 | 13-feb-06 | 7 | 15.4 | 15.4 | 0.288 | 0.215 | 11.300 | 11.433 | 0.353 | 0.354 | 2.975 | 2.724 | | 0.634 | 0.474 | 24.847 | 25.138 | 0.776 | 0.778 | 6.540 | 5.989 |
| rinsing | | | 44.6 | 44.6 | 0.172 | 0.18 | 16.162 | 16.967 | 0.397 | 0.422 | 3.004 | 3.142 | funnel | 0.275 | 0.286 | 25.736 | 27.018 | 0.633 | 0.672 | 4.784 | 5.004 |
| **16/01/2006-13/02/2006** | | | | | | | | | | | | | collector | 0.529 | 0.483 | 25.956 | 25.968 | 0.560 | 0.549 | 5.248 | 4.932 |
| | | | | | | | | | | | | | funnel + collector | 0.804 | 0.769 | 51.692 | 52.986 | 1.192 | 1.221 | 10.032 | 9.936 |
| 13-feb-06 | 20-feb-06 | 7 | 34.4 | 34.8 | 0.143 | 0.164 | 30.447 | 33.151 | 0.257 | 0.304 | 1.763 | 2.114 | | 0.703 | 0.814 | ##### | 164.609 | 1.263 | 1.508 | 8.672 | 10.498 |
| 20-feb-06 | 28-feb-06 | 8 | 13.0 | 13.0 | 0.237 | 0.199 | 15.686 | 15.038 | 0.348 | 0.324 | 1.586 | 1.449 | | 0.385 | 0.322 | 25.489 | 24.347 | 0.565 | 0.524 | 2.578 | 2.345 |
| 28-feb-06 | 6-mar-06 | 6 | 4.9 | 4.9 | 0.498 | 0.648 | 21.241 | 24.593 | 0.458 | 0.479 | 2.505 | 3.359 | | 0.410 | 0.527 | 17.487 | 19.985 | 0.377 | 0.390 | 2.062 | 2.729 |
| 6-mar-06 | 13-mar-06 | 7 | 32.1 | 32.6 | 0.142 | 0.146 | 6.589 | 7.328 | 0.214 | 0.223 | 1.869 | 1.628 | | 0.651 | 0.683 | 30.242 | 34.171 | 0.982 | 1.041 | 8.577 | 7.594 |
| rinsing | | | 25.5 | 25.5 | 0.344 | 0.238 | 28.669 | 30.663 | 0.266 | 0.297 | 3.034 | 3.819 | funnel | 0.313 | 0.216 | 26.086 | 27.901 | 0.242 | 0.270 | 2.761 | 3.475 |
| **13/02/2006-13/03/2006** | | | | | | | | | | | | | collector | 0.537 | 0.586 | 55.747 | 60.778 | 0.797 | 0.866 | 5.472 | 5.792 |
| | | | | | | | | | | | | | funnel + collector | 0.850 | 0.803 | 81.833 | 88.679 | 1.039 | 1.136 | 8.233 | 9.267 |
| 13-mar-06 | 20-mar-06 | 7 | | | | | | | | | | | | | | | | | | | |
| 20-mar-06 | 27-mar-06 | 7 | 20.5 | 20.3 | 0.129 | 0.137 | 4.385 | 5.302 | 0.161 | 0.187 | 0.972 | 1.292 | | 0.377 | 0.397 | 12.862 | 15.411 | 0.473 | 0.542 | 2.850 | 3.755 |
| 27-mar-06 | 3-apr-06 | 7 | 26.3 | 25.9 | 0.179 | 0.180 | 6.329 | 6.973 | 0.173 | 0.208 | 1.882 | 2.132 | | 0.671 | 0.667 | 23.754 | 25.819 | 0.650 | 0.770 | 7.064 | 7.894 |
| 3-apr-06 | 10-apr-06 | 7 | 6.1 | 6.0 | 0.392 | 0.354 | 34.537 | 35.785 | 0.584 | 0.586 | 7.439 | 6.856 | | 0.342 | 0.304 | 30.137 | 30.689 | 0.510 | 0.502 | 6.491 | 5.879 |
| rinsing | | | 25.5 | 25.5 | 0.216 | 0.241 | 36.392 | 39.241 | 0.338 | 0.360 | 2.894 | 3.107 | funnel | 0.196 | 0.219 | 33.114 | 35.706 | 0.307 | 0.328 | 2.633 | 2.827 |
| **13/03/2006-10/04/2006** | | | | | | | | | | | | | collector | 0.464 | 0.456 | 22.251 | 23.973 | 0.544 | 0.605 | 5.469 | 5.843 |
| | | | | | | | | | | | | | funnel + collector | 0.660 | 0.675 | 55.364 | 59.679 | 0.852 | 0.933 | 8.102 | 8.670 |
| 10-apr-06 | 18-apr-06 | 8 | 14.8 | 14.5 | 0.161 | 0.180 | 7.376 | 8.159 | 0.362 | 0.357 | 1.763 | 1.981 | | 0.299 | 0.325 | 13.654 | 14.755 | 0.670 | 0.645 | 3.264 | 3.583 |
| 18-apr-06 | 25-apr-06 | 7 | 0.5 | 0.5 | 1.837 | 1.762 | 84.193 | 83.426 | 2.425 | 2.489 | 37.563 | 38.408 | | 0.129 | 0.117 | 5.899 | 5.541 | 0.170 | 0.165 | 2.632 | 2.551 |
| 25-apr-06 | 2-mai-06 | 7 | 10.7 | 10.5 | 0.469 | 0.515 | 20.956 | 20.579 | 0.849 | 0.818 | 4.635 | 4.647 | | 0.716 | 0.772 | 31.996 | 30.859 | 1.297 | 1.226 | 7.077 | 6.969 |
| 2-mai-06 | 9-mai-06 | 7 | 0.0 | 0.0 | 6.10 | 8.12 | 757 | 975 | 11.3 | 14.7 | 63 | 109 | | 0.017 | 0.011 | 2.066 | 1.331 | 0.031 | 0.020 | 0.172 | 0.149 |
| rinsing | | | 23.8 | 24.7 | 0.455 | 0.459 | 39.711 | 39.693 | 0.842 | 0.800 | 5.402 | 5.777 | funnel | 0.374 | 0.391 | 32.620 | 33.848 | 0.691 | 0.682 | 4.437 | 4.926 |
| **10/04/2006-09/05/2006** | | | | | | | | | | | | | collector | 0.290 | 0.306 | 13.404 | 13.122 | 0.542 | 0.514 | 3.286 | 3.313 |
| | | | | | | | | | | | | | funnel + collector | 0.664 | 0.698 | 46.024 | 46.969 | 1.233 | 1.197 | 7.724 | 8.239 |
| 9-mai-06 | 16-mai-06 | 7 | 9.7 | 10.1 | 0.436 | 0.551 | 19.390 | 21.573 | 0.912 | 0.951 | 5.050 | 5.309 | | 0.604 | 0.793 | 26.888 | 31.034 | 1.265 | 1.369 | 7.002 | 7.637 |
| 16-mai-06 | 23-mai-06 | 7 | 46.6 | 46.5 | 0.100 | 0.097 | 6.218 | 5.391 | 0.198 | 0.183 | 1.070 | 0.993 | | 0.668 | 0.647 | 41.410 | 35.807 | 1.320 | 1.213 | 7.127 | 6.598 |
| 23-mai-06 | 30-mai-06 | 7 | 43.1 | 34.6 | 0.200 | 0.199 | 9.768 | 10.333 | 0.219 | 0.227 | 1.989 | 2.100 | | 1.228 | 0.987 | 60.108 | 51.137 | 1.345 | 1.122 | 12.240 | 10.391 |
| 30-mai-06 | 6-jun-06 | 7 | 2.3 | 1.0 | 0.732 | 0.636 | 79.255 | 86.340 | 0.759 | 0.716 | 13.159 | 11.622 | | 0.240 | 0.087 | 25.925 | 11.784 | 0.248 | 0.098 | 4.304 | 1.586 |
| rinsing | | | 25.1 | 25.2 | 0.359 | 0.345 | 38.223 | 38.707 | 0.386 | 0.358 | 3.277 | 3.162 | funnel | 0.322 | 0.311 | 34.245 | 34.846 | 0.346 | 0.322 | 2.936 | 2.847 |
| **09/05/2006-06/06/2006** | | | | | | | | | | | | | collector | 0.685 | 0.628 | 38.583 | 32.441 | 1.044 | 0.951 | 7.669 | 6.553 |
| | | | | | | | | | | | | | funnel + collector | 1.007 | 0.939 | 72.828 | 67.287 | 1.390 | 1.273 | 10.605 | 9.400 |
| 6-jun-06 | 13-jun-06 | 7 | | | | | | | | | | | | | | | | | | | |
| 13-jun-06 | 20-jun-06 | 7 | 29.2 | 29.4 | 0.154 | 0.157 | 6.70 | 7.19 | 0.378 | 0.374 | 1.591 | 1.462 | | 0.641 | 0.660 | 27.919 | 30.158 | 1.576 | 1.570 | 6.629 | 6.130 |
| 20-jun-06 | 27-jun-06 | 7 | 8.8 | 8.6 | 1.146 | 1.03 | 84.47 | 70.50 | 1.242 | 1.17 | 7.856 | 6.89 | | 1.440 | 1.265 | ##### | 86.601 | 1.560 | 1.437 | 9.871 | 8.464 |
| 27-jun-06 | 4-jul-06 | 7 | | | | | | | | | | | | | | | | | | | |
| rinsing | | | 24.5 | 24.6 | 0.539 | 0.587 | 42.03 | 45.88 | 0.573 | 0.594 | 6.368 | 7.006 | funnel | 0.472 | 0.515 | 36.826 | 40.295 | 0.502 | 0.522 | 5.579 | 6.152 |
| **06/06/2006-04/07/2006** | | | | | | | | | | | | | collector | 1.041 | 0.963 | 67.033 | 58.380 | 1.568 | 1.503 | 8.250 | 7.297 |
| | | | | | | | | | | | | | funnel + collector | 1.513 | 1.478 | ##### | 98.675 | 2.070 | 2.025 | 13.829 | 13.449 |

# A.12 The wet only measurements at Duisburg

| | | | mm | | Concentration Cd | | Pb | | As | | Ni | | | deposition Cd | | Pb | | As | | Ni | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **first day** | **last day** | nr of days | G | H | G | H | G | H | G | H | G | H | | G | H | G | H | G | H | G | H |
| 16-jan-06 | 23-jan-06 | 7 | 14.3 | 13.7 | 0.145 | 0.143 | 6.280 | 6.603 | 0.199 | 0.206 | 1.257 | 1.348 | | 0.296 | 0.279 | 12.809 | 12.897 | 0.406 | 0.403 | 2.563 | 2.632 |
| 23-jan-06 | 30-jan-06 | 7 | 3.69 | 3.82 | 0.360 | 0.357 | 13.509 | 15.455 | 0.509 | 0.561 | 3.065 | 3.215 | | 0.190 | 0.195 | 7.112 | 8.438 | 0.268 | 0.306 | 1.613 | 1.755 |
| 30-jan-06 | 6-feb-06 | 7 | 2.89 | 2.87 | 1.830 | 1.575 | ###### | 86.313 | 1.521 | 1.436 | 15.354 | 14.649 | | 0.755 | 0.645 | 41.479 | 35.361 | 0.627 | 0.588 | 6.335 | 6.001 |
| 6-feb-06 | 13-feb-06 | 7 | 13.9 | 15 | 0.249 | 0.239 | 13.804 | 12.064 | 0.372 | 0.331 | 3.293 | 3.548 | | 0.494 | 0.511 | 27.413 | 25.824 | 0.738 | 0.708 | 6.539 | 7.595 |
| rinsing | | | 16.3 | 16.3 | 0.120 | 0.111 | 11.117 | 10.422 | 0.249 | 0.215 | 1.416 | 1.371 | funnel | 0.070 | 0.065 | 6.474 | 6.069 | 0.145 | 0.125 | 0.825 | 0.799 |
| **16/01/2006-13/02/2006** | | | | | | | | | | | | | wet dep | 0.434 | 0.407 | 22.203 | 20.630 | 0.510 | 0.501 | 4.263 | 4.496 |
| | | | | | | | | | | | | | collector+ funnel | 0.504 | 0.472 | 28.677 | 26.699 | 0.655 | 0.627 | 5.087 | 5.294 |
| 13-feb-06 | 20-feb-06 | 7 | 38.5 | 37.6 | 0.087 | 0.070 | 13.654 | 12.812 | 0.137 | 0.127 | 0.777 | 0.784 | | 0.476 | 0.375 | 75.121 | 68.740 | 0.753 | 0.682 | 4.275 | 4.206 |
| 20-feb-06 | 28-feb-06 | 8 | 12.5 | 11.6 | 0.208 | 0.186 | 14.492 | 12.606 | 0.282 | 0.259 | 1.030 | 0.808 | | 0.326 | 0.269 | 22.696 | 18.246 | 0.442 | 0.375 | 1.613 | 1.170 |
| 28-feb-06 | 6-mar-06 | 6 | 4.79 | 5.17 | 0.383 | 0.303 | 19.358 | 15.171 | 0.372 | 0.328 | 1.766 | 1.463 | | 0.305 | 0.261 | 15.440 | 13.080 | 0.296 | 0.283 | 1.408 | 1.261 |
| 6-mar-06 | 13-mar-06 | 7 | 20.4 | 20.4 | 0.100 | 0.104 | 6.464 | 5.958 | 0.208 | 0.216 | 1.267 | 1.129 | | 0.292 | 0.301 | 18.860 | 17.330 | 0.606 | 0.629 | 3.695 | 3.283 |
| rinsing | | | 16.3 | 16.3 | 0.120 | 0.111 | 6.695 | 7.087 | 0.076 | 0.085 | 0.728 | 0.751 | funnel | 0.070 | 0.065 | 3.899 | 4.127 | 0.044 | 0.049 | 0.424 | 0.438 |
| **13/02/2006-13/03/2006** | | | | | | | | | | | | | wet dep | 0.350 | 0.302 | 33.029 | 29.349 | 0.524 | 0.492 | 2.748 | 2.480 |
| | | | | | | | | | | | | | collector+ funnel | 0.420 | 0.366 | 36.928 | 33.476 | 0.569 | 0.542 | 3.172 | 2.918 |
| 13-mar-06 | 20-mar-06 | 7 | | | | | | | | | | | | | | | | | | | |
| 20-mar-06 | 27-mar-06 | 7 | 24.2 | 23.9 | 0.083 | 0.077 | 2.821 | 2.798 | 0.109 | 0.103 | 0.572 | 0.542 | | 0.287 | 0.262 | 9.733 | 9.564 | 0.377 | 0.353 | 1.973 | 1.854 |
| 27-mar-06 | 3-apr-06 | 7 | 30.7 | 30.7 | 0.087 | 0.123 | 4.771 | 4.896 | 0.123 | 0.135 | 1.170 | 1.217 | | 0.383 | 0.537 | 20.905 | 21.454 | 0.539 | 0.592 | 5.126 | 5.332 |
| 3-apr-06 | 10-apr-06 | 7 | 6.11 | 6.4 | 0.202 | 0.174 | 8.802 | 10.184 | 0.480 | 0.409 | 5.264 | 3.536 | | 0.177 | 0.159 | 7.689 | 9.311 | 0.419 | 0.374 | 4.598 | 3.233 |
| rinsing | | | 16.3 | 16.3 | 0.062 | 0.036 | 10.533 | 5.983 | 0.084 | 0.051 | 0.779 | 0.555 | funnel | 0.036 | 0.021 | 6.134 | 3.484 | 0.049 | 0.030 | 0.454 | 0.323 |
| **13/03/2006-10/04/2006** | | | | | | | | | | | | | wet dep | 0.282 | 0.319 | 12.776 | 13.443 | 0.445 | 0.440 | 3.899 | 3.473 |
| | | | | | | | | | | | | | collector+ funnel | 0.318 | 0.340 | 18.909 | 16.927 | 0.494 | 0.470 | 4.353 | 3.796 |
| 10-apr-06 | 18-apr-06 | 8 | 17.3 | 17.1 | 0.161 | 0.139 | 6.876 | 8.926 | 0.305 | 0.357 | 1.528 | 2.383 | | 0.348 | 0.297 | 14.909 | 19.035 | 0.662 | 0.762 | 3.313 | 5.081 |
| 18-apr-06 | 25-apr-06 | 7 | | | | | | | | | | | | | | | | | | | |
| 25-apr-06 | 2-mai-06 | 7 | 11.8 | 11.7 | 0.235 | 0.241 | 13.726 | 13.656 | 0.489 | 0.496 | 1.602 | 1.723 | | 0.397 | 0.404 | 23.221 | 22.863 | 0.828 | 0.830 | 2.710 | 2.885 |
| 2-mai-06 | 9-mai-06 | 7 | | | | | | | | | | | | | | | | | | | |
| rinsing | | | 16.3 | 16.3 | 0.036 | 0.043 | 5.289 | 8.809 | 0.077 | 0.084 | 0.534 | 0.645 | funnel | 0.020 | 0.024 | 2.974 | 4.953 | 0.043 | 0.047 | 0.300 | 0.363 |
| **10/04/2006-09/05/2006** | | | | | | | | | | | | | wet dep | 0.373 | 0.350 | 19.065 | 20.949 | 0.745 | 0.796 | 3.012 | 3.983 |
| | | | | | | | | | | | | | collector+ funnel | 0.393 | 0.374 | 22.039 | 25.902 | 0.788 | 0.843 | 3.312 | 4.346 |
| 9-mai-06 | 16-mai-06 | 7 | 11.5 | 11.7 | 0.254 | 0.242 | 9.697 | 10.584 | 0.523 | 0.546 | 2.095 | 2.104 | | 0.419 | 0.405 | 15.982 | 17.689 | 0.862 | 0.913 | 3.452 | 3.516 |
| 16-mai-06 | 23-mai-06 | 7 | 61.7 | 61.7 | 0.083 | 0.086 | 6.667 | 8.003 | 0.169 | 0.176 | 0.877 | 0.890 | | 0.727 | 0.761 | 58.719 | 70.583 | 1.487 | 1.555 | 7.727 | 7.849 |
| 23-mai-06 | 30-mai-06 | 7 | 46.5 | 48.2 | 0.161 | 0.154 | 7.329 | 7.826 | 0.189 | 0.200 | 1.214 | 1.138 | | 1.071 | 1.060 | 48.699 | 53.843 | 1.255 | 1.376 | 8.067 | 7.832 |
| 30-mai-06 | 6-jun-06 | 7 | 3 | 3.02 | 0.243 | 0.235 | 19.579 | 18.175 | 0.293 | 0.298 | 4.308 | 2.875 | | 0.104 | 0.101 | 8.380 | 7.832 | 0.125 | 0.129 | 1.844 | 1.239 |
| rinsing | | | 16.3 | 16.3 | 0.103 | 0.081 | 12.520 | 12.459 | 0.117 | 0.123 | 1.887 | 1.721 | funnel | 0.060 | 0.047 | 7.291 | 7.256 | 0.068 | 0.072 | 1.099 | 1.002 |
| **09/05/2006-06/06/2006** | | | | | | | | | | | | | wet dep | 0.580 | 0.582 | 32.945 | 37.487 | 0.932 | 0.993 | 5.272 | 5.109 |
| | | | | | | | | | | | | | collector+ funnel | 0.640 | 0.629 | 40.236 | 44.742 | 1.000 | 1.065 | 6.371 | 6.111 |
| 6-jun-06 | 13-jun-06 | 7 | | | | | | | | | | | | | | | | | | | |
| 13-jun-06 | 20-jun-06 | 7 | 35.4 | 33.9 | 0.122 | 0.108 | 4.887 | 4.433 | 0.283 | 0.299 | 1.084 | 1.088 | | 0.619 | 0.523 | 24.745 | 21.452 | 1.434 | 1.449 | 5.486 | 5.266 |
| 20-jun-06 | 27-jun-06 | 7 | 10.1 | 9.95 | 0.639 | 0.646 | 53.663 | 50.715 | 1.034 | 0.915 | 2.835 | 2.573 | | 0.926 | 0.918 | 77.736 | 72.062 | 1.498 | 1.300 | 4.107 | 3.656 |
| 27-jun-06 | 4-jul-06 | 7 | | | | | | | | | | | | | | | | | | | |
| rinsing | | | 16.3 | 16.3 | 0.035 | 0.035 | 4.074 | 3.664 | 0.076 | 0.047 | 4.604 | 0.665 | funnel | 0.021 | 0.021 | 2.372 | 2.134 | 0.044 | 0.027 | 2.681 | 0.387 |
| **06/06/2006-04/07/2006** | | | | | | | | | | | | | wet dep | 0.773 | 0.721 | 51.241 | 46.757 | 1.466 | 1.375 | 4.797 | 4.461 |
| | | | | | | | | | | | | | collector+ funnel | 0.793 | 0.741 | 53.613 | 48.891 | 1.510 | 1.402 | 7.477 | 4.848 |

## A.13 The Bergerhoff measurements at Duisburg

| | | | Deposition flux in µg/m²/d | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | As | | | Ni | | | Cd | | | Pb | | |
| From | To | Days | A | C | D | A | C | D | A | C | D | A | C | D |
| 16.01.2006 | 13.02.2006 | 28 | 1.047 | 1.083 | 1.408 | 26.552 | 23.458 | 23.777 | 0.916 | 0.981 | 0.939 | 47.374 | 44.269 | 44.223 |
| 13.02.2006 | 13.03.2006 | 28 | 1.070 | 1.060 | 1.259 | 16.306 | 17.480 | 18.551 | 0.650 | 0.645 | 0.697 | 52.363 | 52.775 | 51.303 |
| 13.03.2006 | 10.04.2006 | 28 | 0.893 | 1.098 | 1.210 | 14.845 | 14.481 | 14.342 | 0.613 | 0.600 | 0.606 | 45.464 | 45.494 | 42.182 |
| 10.04.2006 | 09.05.2006 | 29 | 1.370 | 1.375 | 1.561 | 14.958 | 16.564 | 16.717 | 0.756 | 0.698 | 0.602 | 39.919 | 42.358 | 43.677 |
| 09.05.2006 | 06.06.2006 | 28 | 2.101 | 2.140 | 1.977 | 16.172 | 19.726 | 19.732 | 0.912 | 1.046 | 0.979 | 62.350 | 66.381 | 61.517 |
| 06.06.2006 | 04.07.2006 | 28 | 1.959 | 2.043 | 1.721 | 19.150 | 21.579 | 17.635 | 0.952 | 1.003 | 0.850 | 64.303 | 64.603 | 59.897 |

## A.14 The bulk measurements at Birkenes

| | | | | | Concentration | | | | | | | | | deposition, µg/m²day | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | mm | | Cd | | Pb | | As | | Ni | | | Cd | | Pb | | As | | Ni | |
| first day | last day | nr of days | E | F | E | F | E | F | E | F | E | F | | E | F | E | F | E | F | E | F |
| 30.10.2006 | 06.11.2006 | 7 | 16.1 | 17.3 | 0.005 | 0.004 | 0.145 | 0.585 | 0.018 | 0.021 | 0.230 | | | 0.011 | 0.010 | 0.334 | 1.444 | 0.043 | 0.051 | 0.529 | 0.529 |
| 06.11.2006 | 13.11.2006 | 7 | 3.2 | 9.6 | 0.061 | 0.015 | 0.914 | 0.924 | 0.082 | 0.116 | 1.080 | 0.255 | | 0.028 | 0.021 | 0.416 | 1.274 | 0.037 | 0.160 | 0.492 | 0.351 |
| 13.11.2006 | 20.11.2006 | 7 | 70.5 | 83.8 | 0.039 | 0.035 | 1.592 | 1.540 | 0.110 | 0.111 | 0.282 | 0.276 | | 0.390 | 0.425 | ##### | ##### | 1.112 | 1.335 | 2.843 | 3.299 |
| 20.11.2006 | 27.11.2006 | 7 | 142.3 | 147.6 | 0.011 | 0.011 | 0.491 | 0.482 | 0.057 | 0.042 | 0.145 | 0.116 | | 0.216 | 0.226 | 9.975 | ##### | 1.155 | 0.890 | 2.947 | 2.454 |
| rinsing | | | 6.4 | 6.4 | 0.005 | 0.001 | 1.718 | 1.593 | 0.012 | 0.014 | 0.035 | 0.035 | funnel | 0.001 | 0.000 | 0.391 | 0.362 | 0.003 | 0.003 | 0.008 | 0.008 |
| **30-okt-06** | **27-nov-06** | | | | | | | | | | | | wet dep | 0.161 | 0.170 | 6.691 | 7.830 | 0.587 | 0.609 | 1.703 | 1.658 |
| | | | | | | | | | | | | | collector+ funnel | 0.162 | 0.171 | 7.082 | 8.192 | 0.590 | 0.612 | 1.711 | 1.666 |
| 27.11.2006 | 04.12.2006 | 7 | 51.8 | 52.1 | 0.038 | 0.041 | 1.778 | 2.024 | 0.109 | 0.113 | 0.313 | 0.419 | | 0.278 | 0.307 | ##### | ##### | 0.805 | 0.838 | 2.316 | 3.118 |
| 04.12.2006 | 11.12.2006 | 7 | 98.2 | 108.0 | 0.005 | 0.005 | 0.306 | 0.289 | 0.053 | 0.032 | 0.110 | 0.132 | | 0.068 | 0.071 | 4.292 | 4.452 | 0.739 | 0.489 | 1.550 | 2.029 |
| 11.12.2006 | 18.12.2006 | 7 | 30.4 | 30.7 | 0.002 | 0.003 | 0.142 | 0.220 | 0.024 | 0.025 | | | | 0.009 | 0.013 | 0.615 | 0.966 | 0.106 | 0.110 | | |
| 18.12.2006 | 25.12.2006 | 7 | 0.1 | 0.1 | | | | | | | | | | | | | | | | | |
| 25.12.2006 | 01.01.2007 | 7 | 74.7 | 74.5 | 0.022 | 0.024 | 1.022 | 1.067 | 0.082 | 0.081 | 0.162 | 0.165 | | 0.234 | 0.256 | ##### | ##### | 0.870 | 0.861 | 1.730 | 1.755 |
| rinsing | | | 6.4 | 6.4 | 0.007 | 0.001 | 1.441 | 1.551 | 0.009 | 0.012 | 0.083 | 0.230 | funnel | 0.002 | 0.000 | 0.328 | 0.353 | 0.002 | 0.003 | 0.019 | 0.052 |
| **27-nov-06** | **01-jan-07** | | | | | | | | | | | | wet dep | 0.147 | 0.162 | 7.244 | 7.962 | 0.630 | 0.574 | 1.865 | 2.301 |
| | | | | | | | | | | | | | collector+ funnel | 0.149 | 0.162 | 7.572 | 8.315 | 0.632 | 0.577 | 1.884 | 2.353 |
| 01.01.2007 | 08.01.2007 | 7 | 28.1 | 25.8 | 0.011 | 0.010 | 0.884 | 0.890 | 0.072 | 0.063 | 0.184 | 0.201 | | 0.044 | 0.037 | 3.551 | 3.280 | 0.290 | 0.231 | 0.738 | 0.742 |
| 08.01.2007 | 15.01.2007 | 7 | 79.6 | 77.2 | 0.005 | 0.005 | 0.235 | 0.235 | 0.056 | 0.048 | 0.103 | 0.094 | | 0.059 | 0.053 | 2.673 | 2.596 | 0.642 | 0.531 | 1.174 | 1.036 |
| 15.01.2007 | 22.01.2007 | 7 | 51.2 | 49.7 | 0.005 | 0.006 | 0.261 | 0.294 | 0.071 | 0.067 | 0.131 | 0.130 | | 0.035 | 0.040 | 1.911 | 2.090 | 0.517 | 0.475 | 0.960 | 0.925 |
| 22.01.2007 | 29.01.2007 | 7 | 0.8 | 0.8 | 0.011 | 0.012 | 0.395 | 0.593 | 0.057 | 0.044 | | 0.600 | | 0.001 | 0.001 | 0.043 | 0.070 | 0.006 | 0.005 | 0.071 | 0.071 |
| 29.01.2007 | 05.02.2007 | 7 | 0.2 | 0.1 | | | | | | | | | | | | | | | | | |
| rinsing | | | 6.4 | 6.4 | 0.002 | 0.002 | 1.010 | 0.709 | 0.036 | 0.029 | 0.131 | 0.078 | funnel | 0.001 | 0.000 | 0.230 | 0.161 | 0.008 | 0.007 | 0.030 | 0.018 |
| **01-jan-07** | **05-feb-07** | | | | | | | | | | | | wet dep | 0.035 | 0.033 | 2.045 | 2.009 | 0.364 | 0.311 | 0.736 | 0.694 |
| | | | | | | | | | | | | | collector+ funnel | 0.035 | 0.033 | 2.275 | 2.170 | 0.372 | 0.317 | 0.766 | 0.711 |
| 05.02.2007 | 12.02.2007 | 7 | 15.9 | 16.1 | 0.022 | 0.022 | 0.695 | 0.638 | 0.087 | 0.079 | 0.326 | 0.166 | | 0.050 | 0.051 | 1.574 | 1.463 | 0.197 | 0.181 | 0.740 | 0.380 |
| 12.02.2007 | 19.02.2007 | 7 | 8.2 | 7.8 | 0.061 | 0.066 | 2.883 | 2.389 | 0.471 | 0.497 | 0.582 | 0.401 | | 0.071 | 0.074 | 3.371 | 2.674 | 0.551 | 0.557 | 0.681 | 0.449 |
| 19.02.2007 | 26.02.2007 | 7 | 68.2 | 65.2 | 0.026 | 0.024 | 0.939 | 0.900 | 0.119 | 0.084 | 0.267 | 0.171 | | 0.254 | 0.225 | 9.151 | 8.373 | 1.158 | 0.782 | 2.599 | 1.593 |
| 26.02.2007 | 05.03.2007 | 7 | 27.6 | 25.2 | 0.014 | 0.014 | 1.004 | 0.968 | 0.083 | 0.093 | 0.324 | 0.345 | | 0.054 | 0.050 | 3.965 | 3.480 | 0.326 | 0.333 | 1.278 | 1.238 |
| rinsing | | | 6.4 | 6.4 | 0.002 | 0.003 | 0.970 | 0.827 | 0.035 | 0.034 | 0.229 | 0.744 | funnel | 0.000 | 0.001 | 0.221 | 0.188 | 0.008 | 0.008 | 0.052 | 0.169 |
| **05-feb-07** | **05-mar-07** | | | | | | | | | | | | wet dep | 0.107 | 0.100 | 4.515 | 3.998 | 0.558 | 0.463 | 1.324 | 0.915 |
| | | | | | | | | | | | | | collector+ funnel | 0.108 | 0.101 | 4.736 | 4.186 | 0.566 | 0.471 | 1.377 | 1.084 |
| 05.03.2007 | 12.03.2007 | 7 | 62.2 | 62.2 | 0.025 | 0.028 | 0.880 | 0.894 | 0.083 | 0.080 | 0.625 | 0.232 | | 0.225 | 0.247 | 7.818 | 7.945 | 0.736 | 0.709 | 5.550 | 2.064 |
| 12.03.2007 | 19.03.2007 | 7 | 17.7 | 19.2 | 0.003 | 0.006 | 0.204 | 0.232 | 0.032 | 0.062 | 0.268 | 0.150 | | 0.008 | 0.017 | 0.516 | 0.637 | 0.081 | 0.170 | 0.679 | 0.412 |
| 19.03.2007 | 26.03.2007 | 7 | 1.1 | 1.6 | 0.088 | 0.045 | 0.952 | 1.056 | 0.154 | 0.130 | 1.126 | 0.322 | | 0.014 | 0.010 | 0.156 | 0.235 | 0.025 | 0.029 | 0.184 | 0.072 |
| 26.03.2007 | 02.04.2007 | 7 | 0.2 | 0.1 | | | | | | | | | | | | | | | | | |
| rinsing | | | 6.4 | 6.4 | 0.013 | 0.015 | 1.198 | 1.486 | 0.042 | 0.036 | 0.369 | 0.161 | funnel | 0.003 | 0.004 | 0.273 | 0.338 | 0.010 | 0.008 | 0.084 | 0.037 |
| **05-mar-07** | **02-apr-07** | | | | | | | | | | | | wet dep | 0.082 | 0.091 | 2.830 | 2.939 | 0.281 | 0.303 | 2.138 | 0.849 |
| | | | | | | | | | | | | | collector+ funnel | 0.085 | 0.095 | 3.103 | 3.277 | 0.291 | 0.311 | 2.222 | 0.886 |
| 02.04.2007 | 09.04.2007 | 7 | 6.5 | 6.8 | 0.021 | 0.032 | 0.726 | 1.102 | 0.063 | 0.078 | 0.554 | 0.468 | | 0.019 | 0.031 | 0.671 | 1.078 | 0.058 | 0.077 | 0.512 | 0.458 |
| 09.04.2007 | 16.04.2007 | 7 | 0.6 | 0.7 | 0.085 | 0.027 | 1.005 | 0.602 | 0.249 | 0.184 | 0.986 | 0.257 | | 0.007 | 0.003 | 0.087 | 0.060 | 0.022 | 0.018 | 0.085 | 0.026 |
| 16.04.2007 | 23.04.2007 | 7 | 10.8 | 11.2 | 0.034 | 0.090 | 1.078 | 0.814 | 0.237 | 0.164 | 1.780 | 0.600 | | 0.052 | 0.052 | 1.664 | 1.664 | 0.365 | 0.365 | 2.748 | 2.748 |
| 23.04.2007 | 01.05.2007 | 8 | 9.3 | 8.6 | 0.021 | 0.026 | 0.902 | 0.757 | 0.147 | 0.140 | 0.561 | 0.397 | | 0.024 | 0.028 | 1.048 | 0.814 | 0.171 | 0.151 | 0.652 | 0.427 |
| rinsing | | | 6.4 | 6.4 | 0.020 | 0.022 | 1.398 | 1.196 | 0.035 | 0.071 | 0.365 | 0.388 | funnel | 0.004 | 0.005 | 0.307 | 0.263 | 0.008 | 0.016 | 0.080 | 0.085 |
| **02-apr-07** | **01-mai-07** | | | | | | | | | | | | wet dep | 0.026 | 0.029 | 0.867 | 0.904 | 0.154 | 0.153 | 0.999 | 0.915 |
| | | | | | | | | | | | | | collector+ funnel | 0.030 | 0.034 | 1.174 | 1.167 | 0.162 | 0.168 | 1.080 | 1.000 |

## A.15 The wet only measurements at Birkenes

| | | | mm | | | Concentration | | | | | | | | | Deposition | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | Cd | | Pb | | As | | Ni | | | Cd | | Pb | | As | | Ni | | |
| **first day** | **last day** | nr of days | G | H | bulk mm | G | H | G | H | G | H | G | H | | G | H | G | H | G | H | G | H | |
| 30.10.06 | 13.11.06 | 14 | 25.38 | 21.86 | 16.70 | 0.016 | | 0.823 | | 0.036 | | 0.279 | | | 0.028 | 0.028 | 1.491 | 1.491 | 0.065 | 0.065 | 0.506 | 0.506 | overflow, used bulk mm |
| 13.11.06 | 20.11.06 | 7 | 59.22 | 59.88 | 77.17 | 0.037 | 0.045 | 1.708 | 1.834 | 0.103 | 0.140 | 0.505 | 0.415 | | 0.402 | 0.493 | 18.825 | 20.212 | 1.131 | 1.544 | 5.569 | 4.574 | overflow, used bulk mm |
| 20.11.06 | 23.11.06 | 3 | 59.82 | 58.28 | | 0.013 | 0.015 | 0.510 | 0.501 | 0.057 | 0.076 | 0.232 | 0.230 | | 0.265 | 0.326 | 10.736 | 10.738 | 1.206 | 1.621 | 4.880 | 4.930 | overflow, used bulk mm |
| 23.11.06 | 25.11.06 | 2 | 58.46 | 58.58 | 142.35 | 0.016 | 0.013 | 0.459 | 0.501 | 0.048 | 0.049 | 0.184 | 0.173 | | 0.456 | 0.384 | 13.414 | 14.677 | 1.391 | 1.421 | 5.372 | 5.073 | |
| 25.11.06 | 27.11.06 | 2 | 20.76 | 19.52 | | 0.013 | 0.015 | 0.645 | 0.677 | 0.051 | 0.051 | 0.287 | 0.274 | | 0.130 | 0.145 | 6.697 | 6.603 | 0.527 | 0.493 | 2.975 | 2.671 | |
| rinsing | | | 4.00 | 4.00 | | 0.002 | 0.002 | 1.351 | 1.302 | 0.004 | 0.009 | 0.151 | 0.095 | funnel | 0.001 | 0.001 | 0.386 | 0.372 | 0.001 | 0.003 | 0.043 | 0.027 | |
| **30.10.06** | **27.11.06** | | | | | | | | | | | | | wet dep | 0.256 | 0.275 | 10.233 | 10.744 | 0.864 | 1.029 | 3.860 | 3.551 | |
| | | | | | | | | | | | | | | collector+ funnel | 0.257 | 0.276 | 10.619 | 11.116 | 0.865 | 1.031 | 3.903 | 3.578 | |
| 27.11.06 | 04.12.06 | 7 | 48.58 | 49.04 | 51.97 | 0.041 | 0.042 | 1.691 | 1.769 | 0.111 | 0.128 | 0.486 | 0.533 | | 0.287 | 0.295 | 11.736 | 12.391 | 0.767 | 0.897 | 3.374 | 3.737 | |
| 04.12.06 | 07.12.06 | 3 | 54.90 | 51.86 | 103.11 | 0.010 | 0.010 | 0.424 | 0.461 | 0.058 | 0.071 | 0.207 | 0.222 | | 0.183 | 0.171 | 7.767 | 7.969 | 1.060 | 1.222 | 3.788 | 3.829 | |
| 07.12.06 | 11.12.06 | 4 | 49.58 | 47.10 | | 0.003 | 0.006 | 0.282 | 0.285 | 0.049 | 0.046 | 0.231 | | | 0.040 | 0.066 | 3.493 | 3.358 | 0.610 | 0.542 | 2.863 | 2.863 | |
| 11.12.06 | 18.12.06 | 7 | 29.00 | 27.24 | 30.56 | 0.003 | 0.004 | 0.172 | 0.218 | 0.035 | 0.040 | 0.178 | 0.255 | | 0.010 | 0.015 | 0.712 | 0.847 | 0.146 | 0.155 | 0.737 | 0.991 | |
| 18.12.06 | 25.12.06 | 7 | | | | | | | | | | | | | | | | | | | | | |
| 25.12.06 | 01.01.07 | 7 | 59.96 | 59.84 | 74.63 | 0.025 | 0.032 | 1.083 | 1.052 | 0.080 | 0.095 | 0.228 | 0.232 | | 0.270 | 0.338 | 11.548 | 11.211 | 0.850 | 1.007 | 2.433 | 2.469 | overflow, used bulk mm |
| rinsing | | | 4.00 | 4.00 | | 0.002 | 0.021 | 2.046 | 2.398 | 0.004 | 0.008 | 0.041 | 0.124 | funnel | 0.000 | 0.003 | 0.327 | 0.384 | 0.001 | 0.001 | 0.007 | 0.020 | |
| **27.11.06** | **01.01.07** | | | | | | | | | | | | | wet dep | 0.158 | 0.177 | 7.051 | 7.155 | 0.686 | 0.765 | 2.639 | 2.778 | |
| | | | | | | | | | | | | | | collector+ funnel | 0.158 | 0.180 | 7.378 | 7.539 | 0.687 | 0.766 | 2.646 | 2.798 | |
| 01.01.07 | 08.01.07 | 7 | 28.92 | 18.62 | 26.95 | 0.011 | 0.014 | 0.729 | 0.826 | 0.078 | 0.076 | 0.342 | 0.406 | | 0.047 | 0.036 | 3.011 | 2.198 | 0.324 | 0.203 | 1.412 | 1.079 | |
| 08.01.07 | 15.01.07 | 7 | 57.94 | 57.30 | 78.44 | 0.007 | 0.008 | 0.237 | 0.296 | 0.052 | 0.059 | 0.137 | 0.222 | | 0.080 | 0.087 | 2.659 | 3.318 | 0.577 | 0.659 | 1.537 | 2.484 | overflow, used bulk mm |
| 15.01.07 | 22.01.07 | 7 | 50.48 | | 50.45 | 0.006 | | 0.279 | | 0.062 | | 0.142 | | | 0.043 | 0.043 | 2.008 | 2.008 | 0.450 | 0.450 | 1.026 | 1.026 | |
| 22.01.07 | 29.01.07 | 7 | | | | | | | | | | | | | | | | | | | | | |
| 29.01.07 | 05.02.07 | 7 | | | | | | | | | | | | | | | | | | | | | |
| rinsing | | | 4.00 | 4.00 | | 0.004 | 0.013 | 1.802 | 1.719 | 0.011 | 0.015 | 0.151 | 0.291 | funnel | 0.001 | 0.002 | 0.257 | 0.246 | 0.002 | 0.002 | 0.022 | 0.042 | |
| **01.01.07** | **05.02.07** | | | | | | | | | | | | | wet dep | 0.056 | 0.055 | 2.560 | 2.508 | 0.450 | 0.437 | 1.325 | 1.530 | |
| | | | | | | | | | | | | | | collector+ funnel | 0.057 | 0.057 | 2.817 | 2.754 | 0.452 | 0.439 | 1.347 | 1.572 | |
| 05.02.07 | 12.02.07 | 7 | 14.38 | | 15.96 | 0.020 | | 0.525 | | 0.090 | | 0.196 | | | 0.041 | | 1.078 | | 0.184 | | 0.403 | | |
| 12.02.07 | 19.02.07 | 7 | 6.50 | | 8.01 | 0.079 | | 2.571 | | 0.584 | | 0.457 | | | 0.073 | | 2.388 | | 0.542 | | 0.424 | | |
| 19.02.07 | 24.02.07 | 5 | 57.00 | | 66.70 | 0.058 | | 1.863 | | 0.223 | | 0.175 | | | 0.656 | | 21.237 | | 2.539 | | 1.993 | | |
| 24.02.07 | 26.02.07 | 2 | 42.16 | | | 0.024 | | 0.816 | | 0.078 | | 0.091 | | | 0.508 | | 17.203 | | 1.638 | | 1.927 | | |
| 26.02.07 | 05.03.07 | 7 | 21.00 | | 26.40 | 0.014 | | 0.900 | | 0.062 | | 0.236 | | | 0.041 | | 2.699 | | 0.185 | | 0.707 | | |
| rinsing | | | 4.00 | | | 0.007 | | 2.312 | | 0.025 | | 0.216 | | funnel | 0.001 | | 0.440 | | 0.005 | | 0.041 | | |
| **05.02.07** | **05.03.07** | | | | | | | | | | | | | wet dep | 0.264 | | 8.921 | | 1.018 | | 1.091 | | |
| | | | | | | | | | | | | | | collector+ funnel | 0.265 | | 9.361 | | 1.022 | | 1.132 | | |
| 05.03.07 | 06.03.07 | 1 | 48.88 | | 62.20 | 0.035 | | 0.685 | | 0.092 | | 0.325 | | | 1.725 | | 33.473 | | 4.497 | | 15.896 | | |
| 06.03.07 | 12.03.07 | 6 | 15.24 | | | 0.017 | | 0.978 | | 0.057 | | 0.286 | | | 0.043 | | 2.484 | | 0.145 | | 0.727 | | |
| 12.03.07 | 19.03.07 | 7 | 18.76 | | 18.49 | 0.005 | | 0.232 | | 0.047 | | 0.212 | | | 0.013 | | 0.621 | | 0.126 | | 0.568 | | |
| 19.03.07 | 26.03.07 | 7 | 2.22 | | 1.35 | 0.059 | | 1.332 | | 0.110 | | 0.280 | | | 0.019 | | 0.422 | | 0.035 | | 0.089 | | |
| 26.03.07 | 02.04.07 | 7 | | | 0.11 | | | | | | | | | | | | | | | | | | |
| rinsing | | | 4.00 | | | 0.008 | | 2.222 | | 0.006 | | 0.112 | | funnel | 0.001 | | 0.329 | | 0.001 | | 0.017 | | |
| **05.03.07** | **02.04.07** | | | | | | | | | | | | | wet dep | 0.450 | | 9.250 | | 1.201 | | 4.320 | | |
| | | | | | | | | | | | | | | collector+ funnel | 0.451 | | 9.579 | | 1.201 | | 4.336 | | |
| 02.04.07 | 09.04.07 | 7 | 6.48 | | 6.66 | 0.010 | | 0.180 | | 0.052 | | 0.114 | | | 0.009 | | 0.166 | | 0.048 | | 0.106 | | |
| 09.04.07 | 16.04.07 | 7 | 2.30 | | 0.65 | 0.044 | | 0.306 | | 0.098 | | 0.196 | | | 0.014 | | 0.101 | | 0.032 | | 0.064 | | |
| 16.04.07 | 23.04.07 | 7 | 11.18 | | 11.03 | 0.025 | | 0.611 | | 0.087 | | 0.465 | | | 0.040 | | 0.976 | | 0.139 | | 0.743 | | |
| 23.04.07 | 01.05.07 | 8 | 8.64 | | 8.95 | 0.025 | | 0.806 | | 0.117 | | 0.485 | | | 0.027 | | 0.870 | | 0.126 | | 0.524 | | |
| rinsing | | | 4.00 | | | 0.013 | | 1.558 | | 0.010 | | 0.152 | | funnel | 0.002 | | 0.215 | | 0.001 | | 0.021 | | |
| **02.04.07** | **01.05.07** | | | | | | | | | | | | | wet dep | 0.108 | | 2.339 | | 0.309 | | 1.155 | | |
| | | | | | | | | | | | | | | collector+ funnel | 0.110 | | 2.553 | | 0.311 | | 1.176 | | |

## A.16 The bergerhoff measurements at Birkenes

| | | | Deposition flux in µg/m²/d | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | **As** | | | **Ni** | | | **Cd** | | | **Pb** | | |
| From | To | Days | EMD | EMD | EMD | EMD | EMD | EMD | EMD | EMD | EMD | EMD | EMD | EMD |
| 01.11.2006 | 01.12.2006 | 30 | 0.587 | 0.589 | 0.670 | 2.310 | 2.496 | 2.477 | 0.240 | 0.231 | 0.249 | 6.981 | 4.516 | 3.898 |
| 01.12.2006 | 01.01.2007 | 31 | 0.603 | 0.389 | 0.184 | 1.379 | 1.532 | 0.733 | 0.129 | 0.122 | 0.076 | 2.448 | 2.239 | 0.931 |
| 01.01.2007 | 01.02.2007 | 31 | 0.451 | 0.647 | 0.486 | 0.668 | 1.162 | 1.650 | 0.049 | 0.109 | 0.129 | 0.785 | 2.271 | 0.780 |
| 01.02.2007 | 01.03.2007 | 28 | 0.131 | 0.182 | 0.116 | 0.438 | 0.546 | 0.396 | 0.066 | 0.068 | 0.063 | 1.549 | 1.612 | 0.942 |
| 01.03.2007 | 01.04.2007 | 31 | 0.141 | 0.201 | 0.253 | 0.855 | 0.820 | 0.835 | 0.092 | 0.081 | 0.091 | 2.254 | 1.431 | 1.549 |
| 01.04.2007 | 01.05.2007 | 30 | 0.088 | 0.123 | 0.049 | 0.469 | 0.601 | 0.483 | 0.032 | 0.026 | 0.031 | 0.581 | 1.100 | 0.724 |