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Aerosol and volatile emissions control in an amine-based CO₂ capture plant

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Abstract

This work reports on an experimental campaign measuring particles in the flue gas of the waste-to-energy plant of Twence. The plant is equipped with a CO₂ capture pilot facility, which uses a brownian demister unit (BDU) as means of separating particles and avoiding amine emissions. Measurements were performed in July 2022 at the inlet and outlet of the BDU, as well as at the outlet of the absorber water wash. This campaign is compared to results from a previous campaign, performed in March 2021, right after the installation of the BDU on-site.

In the July 2022 campaign, the total mass of particles entering the BDU was of the order of magnitude 10¹ mg/m³. At this relatively low number, when the BDU was by-passed, no increase in the emissions were observed. On the other hand, the March 2021 campaign had a much higher mass of particles entering the BDU, with order of magnitude 10³ mg/m³. When by-passing the BDU, an immediate increase in emissions was observed: from ca. 2.5 to 750 mg/Nm³.

It is confirmed that the BDU is an efficient technique for lowering aerosol-based emissions, with separation efficiency above 99.8% measured in the campaign of March 2021, and even higher, 99,98%, in the campaign of July 2022.

Keywords: emissions; amine; CO₂ capture; brownian demister unit

1. Introduction

Post-combustion CO₂ capture (PCC) is critical for reduction of greenhouse gas emissions, particularly in hard-to-decarbonize industrial sectors, such as Waste-to-Energy (WtE). Amine-based technology has been implemented for capturing CO₂ from several types of flue gas sources. CO₂ capture using aqueous 30wt% monoethanolamine (MEA) is considered as the benchmark technology [1], and is also being used at pilot scale at Twence, a WtE plant located in the east of The Netherlands. In the WtE context, PCC can lead to negative emissions since most of the CO₂ captured is biogenic [2]. This form of BECCS (bio-energy with CO₂ capture and storage) can contribute to decarbonizing the electricity grid. Also, captured biogenic CO₂ can be a feedstock for future fuels and chemicals, contributing to the defossilisation of the chemical sector [3]. Having these applications in mind, Twence has been a first mover in

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accelerating WtE PCC. In 2022, it has started to build a CO₂ capture plant with capacity to produce 100 kilotonne of CO₂ per year (ktpa).

The CO₂ capture pilot used in this study was commissioned in 2014 [4], and has been operational since then. The pilot treats ca. 1.5 vol% of the flue gas from Twence's third waste incinerator line (ca. 3300 Nm³/h), and produces up to 500 kg/h of CO₂. Part of the CO₂ is used to produce sodium bicarbonate, which is directly used in the flue gas treatment of the same incinerator line. After a modification in 2019 the CO₂ can also be purified, liquified, and sold for external use (e.g., in the horticulture sector). After replacement of the original solvent in 2019 it is mostly operated with 30 wt% monoethanolamine (MEA). However, during the tests reported in this paper (performed in July 2022), the plant was operational with the commercial APBS-CDRMax solvent from Carbon Clean, as part of the research project NEWEST-CCUS.

There are economic and environmental challenges associated with the wide spread implementation of PCC. Emission of amine solvents not only increases the environmental impact of PCC, but also leads to higher operating costs. The volatile nature of amines contributes to emissions and observed MEA emissions in the treated flue gas at pilot plant operations are typically either below 10 mg/Nm³ or in the range of 100–1500 mg/Nm³ [5]. The high emission range is caused by aerosol-based emissions, and is significantly higher than e.g. the emission limit of 15 mg/Nm³ proposed at the TCM plant [6]. Traditionally, water wash and demisters are used as emission management techniques. But these counter measures are not sufficient for mitigating aerosol-based amine emissions, which are caused by the presence of aerosol particles in the gas, with diameter below 5 µm [5]. The most mature technology (TRL7) for controlling aerosol-based amine emissions is the Brownian Demister Unit (BDU), a filter capable of removing small particles from the flue gas [5]. While a BDU was used at different campaigns at the TCM pilot plant, the publications on those campaign focus on reporting the efficiency in particle removal, but fail to report on the impact on amine emissions reduction [6], [7].

Since 2021, a BDU is also operated at the Twence pilot plant. Initial results measured shortly after the commissioning of the filter confirmed a dramatic decrease in MEA emissions reduction – from 750 mg/Nm³ to 2.7 mg/Nm³, following a particle separation efficiency above 99% [8]. Twence and TNO are both partners of the 3-years SCOPE transnational project (Sustainable OPERation of post-combustion Capture plants, ACT 3 Project No 327341). Within the project, TNO will perform several tests at the Twence pilot plant facility, to generate more data on volatile and aerosol-based emissions, as well as on the long-term performance of the BDU. This paper presents a first set of data, obtained during measurements at the Twence pilot performed in July 2022, which focused on the efficiency of particle removal by the BDU.

2. Methodology

The particle number concentration and size distribution were measured using an the electrical low-pressure impactor (ELPI). A Dekati High-Temperature ELPI®+ was used, which allows particles measurements at elevated temperature of up to 180°C. The equipment is shown in Figure 1, with the heated gas sampling probe connected to a flue gas line at the Twence pilot. The ELPI measures particles from 6 nm to 5,44 µm. These particles are collected in impactors, and the number of particles per impactor is quantified. Each impactor collects particles of a given diameter range, and this allows for characterizing the particle number and size distribution.

The particulate matter measurements are performed by connecting a gas sampling line to the desired gas sampling points. The nozzle in the sampling line is chosen based on the gas speed, so that isokinetic sampling is attempted (i.e., the gas velocity inside the piping is the same as the gas velocity inside the probe). Particle losses are avoided by having as short and straight sampling lines as possible, and by adjusting the pressure (and thus the gas flowrate) so that laminar flow is ensured, avoiding turbulent impaction. The sample lines are of conductive material to avoid electrostatic losses, and the temperature is controlled at sample temperature (or slightly higher) as the flue gas at the sample point to avoid thermophoretic losses.

The BDU filter is installed at the Twence pilot downstream the quench, as schematically shown in Figure 2. In the quench, the temperature of the gas is rapidly reduced (from ca. 160°C to ca. 40°C), and the formation of a sulfuric acid mist may occur, depending on the SO_x content in the gas. The BDU separates the aerosols, which are collected in a condensate vessel at the bottom of the candle filters.

The particle measurements were performed at 3 different sampling points, which are indicated in Figure 2 by the numbered circles.



Figure 1. ELPI connected to a flue gas line at Twence's CO₂ capture pilot plant

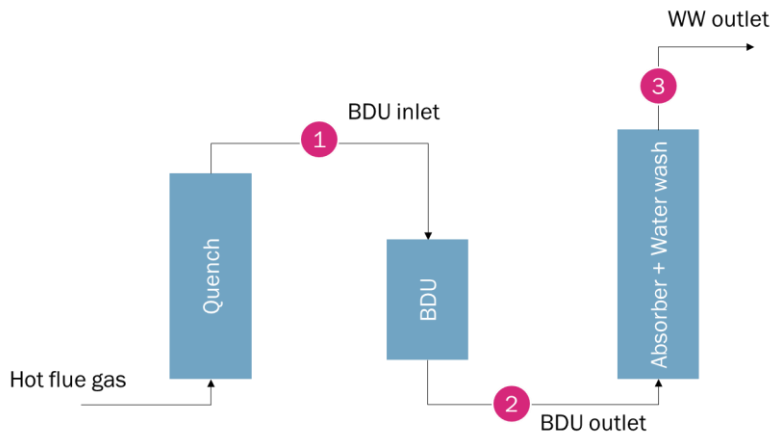


Figure 2. Schematic representation of location of sampling points used for Twence's CO₂ capture pilot plant

3. Results and Discussions

All particle size distribution graphs are presented in log-log scale. A continuous line indicates the distribution, whereas the data points indicate the measurements from each stage in the ELPI. The y-axis presents the particle number

distribution in a concentration unit (number of particles per cubic centimetre of gas), whereas the x-axis gives the particle sizes in μm . This particle size refers to the median diameter or medium value of particle size distribution, commonly represented by D50. This diameter represents the value of the particle diameter at 50% in the cumulative distribution. For example, if an ELPI stage has $D50=5.4\ \mu\text{m}$, then 50% of the particles in that stage count are larger than $5.4\ \mu\text{m}$, and 50% smaller.

Each measurement series is named to indicate which stream was being measured, and when the measurements took place. The name of the series starts with SX – indicating the sample point X (1 to 3, see Figure 2). Next, the date (either 20/07 or 21/07) and time (e.g., 1500 – to indicate 15h00) when the measurement started are given.

The particle size distributions measured at the BDU inlet (sampling port 1) are given in Figure 3. A total of 5 measurements were carried out, with total average particle number $1.73 \cdot 10^7$, and standard deviation $1.21 \cdot 10^6$. The standard deviation is 7% of the average, indicating good repeatability of the measurements taken.

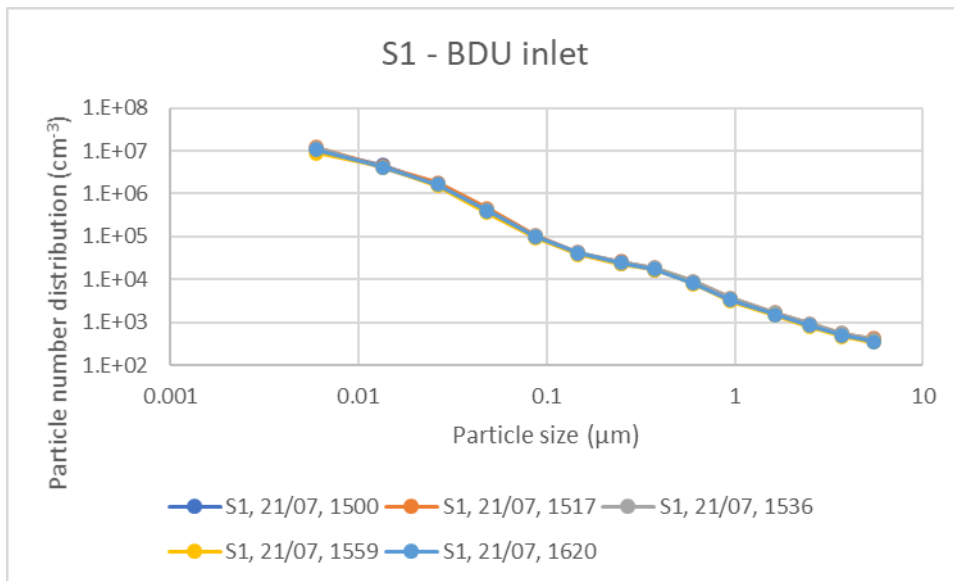


Figure 3 – Particle size distribution measured with the ELPI at sampling port 1 (BDU inlet)

The particle size distributions measured at the BDU outlet (sampling port 2) are given in Figure 4. A total of 5 measurements were carried out, with total average particle number $3.47 \cdot 10^3$, and standard deviation $5.20 \cdot 10^2$. The standard deviation is 15% of the average, indicating good repeatability of the measurements taken.

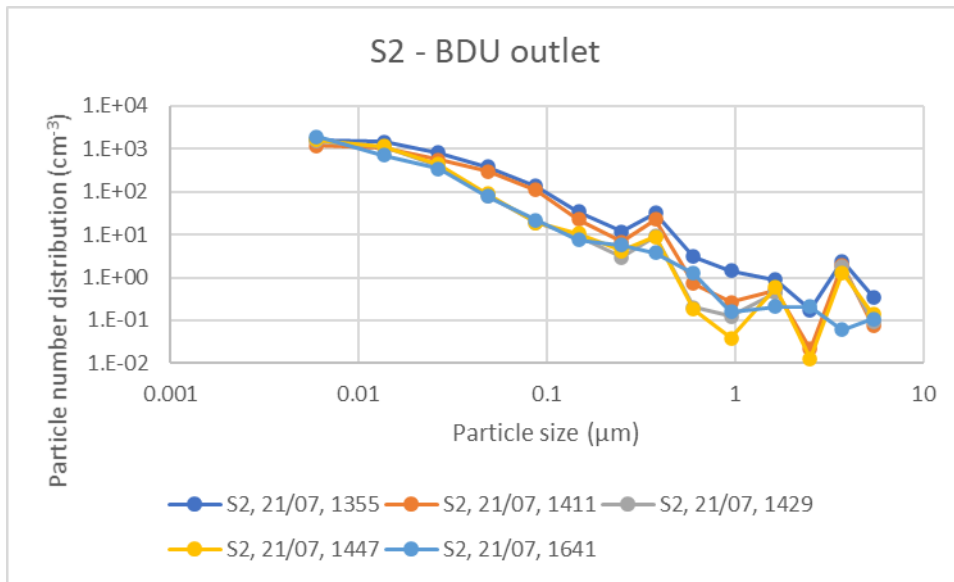


Figure 4 – Particle size distribution measured with the ELPI at sampling port 2 (BDU outlet)

The particle size distributions measured at the WW outlet (sampling port 3) are given in Figure 5 and Figure 6. The five measurements shown in in Figure 5 were performed while the BDU was online. For the measurements shown in Figure 6, the BDU was by-passed. The total average particle number measured while the BDU was online was $4.60 \cdot 10^4$, with a standard deviation of $5.47 \cdot 10^4$. The standard deviation is 119% of the average, indicating bad repeatability of the measurements taken. It could be that process variations caused a disturbance in the particle numbers while the measurements were taken. When the BDU was by-passed, the total average particle number measured increased to $1.27 \cdot 10^7$, with a standard deviation of $7.69 \cdot 10^5$. The standard deviation is 6% of the average, indicating good repeatability of the measurements taken.

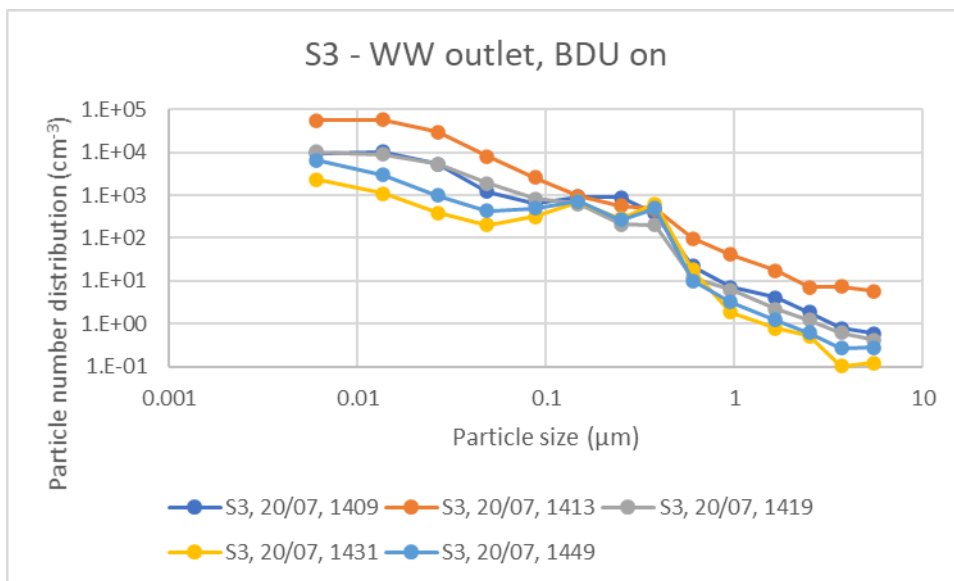


Figure 5 – Particle size distribution measured with the ELPI at sampling port 3 (water wash outlet), while the BDU was online

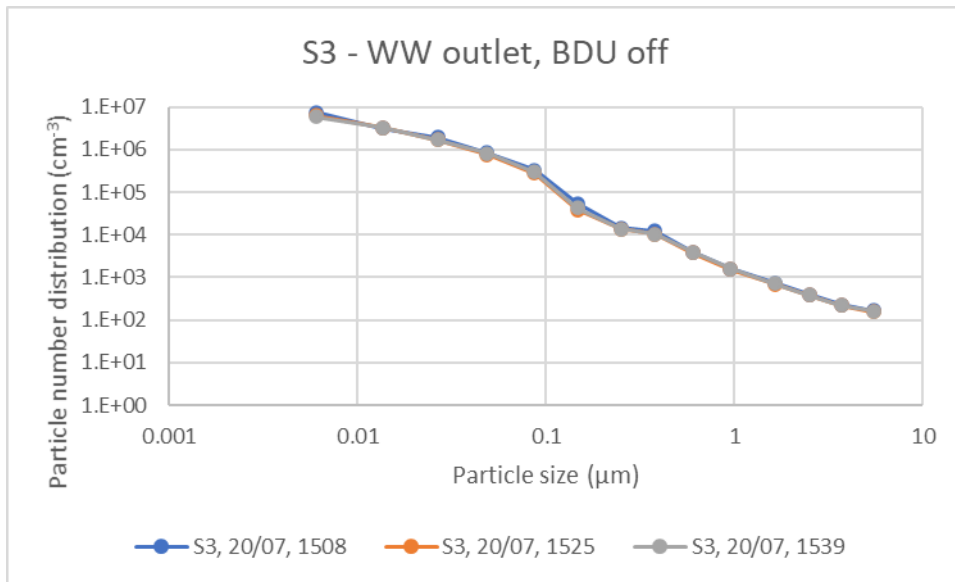


Figure 6 – Particle size distribution measured with the ELPI at sampling port 3 (water wash outlet), while the BDU was bypassed

Figure 7 shows the average particle number distributions. It allows to visualize the great difference between the BDU inlet (S1) and outlet (S2). The BDU separates 99.98% of the particles, considering the average concentration measured at the inlet and the outlet. A sustained particle growth seems to take place within the absorber and water wash (compare S2 to S3 with BDU on). These results will be further analysed within the SCOPE project, and will be used to update and validate the existing model for aerosol particle growth [9].

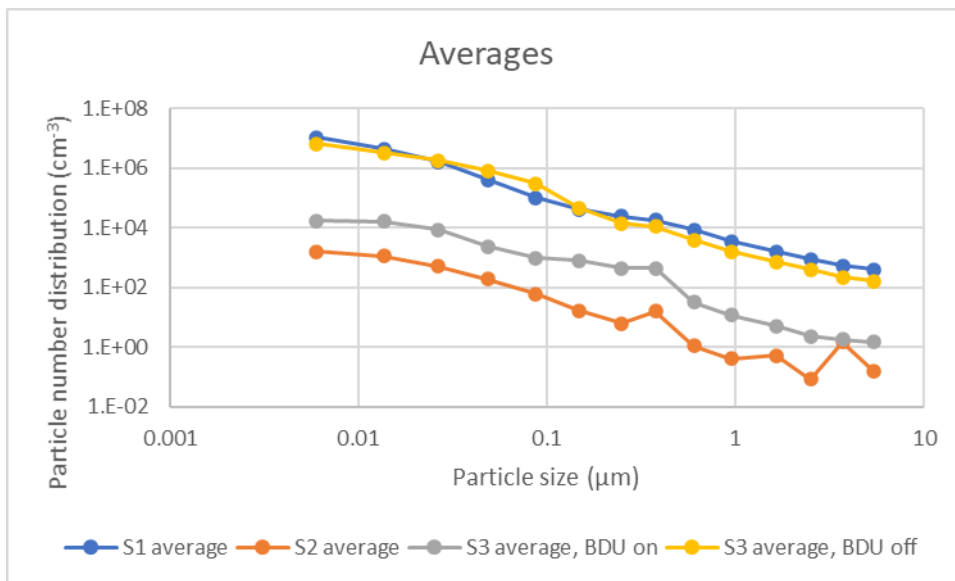


Figure 7 – Average particle size distribution measured with the ELPI at sampling ports 1 to 3

When comparing the water wash outlet (S3) when the BDU is online or by-passed, it is clear that the BDU greatly avoids particle emissions to the atmosphere – particle number concentrations dropped from 10^7 to 10^5 (see Figure 7). The relevance of this towards emissions becomes clear when estimating the total mass of the particles. This is done by considering that all particles are spheric with density of 1000 kg/m^3 , according to Eq. 1. Unit corrections are applied to present the mass in mg/m^3 , which is the unit of measure when FTIR equipment are used to evaluate emissions.

$$M \left(\frac{\text{mg}}{\text{m}^3} \right) = \sum_i PNC_i \left(\frac{1}{\text{cm}^3} \right) * 10^6 \left(\frac{\text{cm}^3}{\text{m}^3} \right) * \frac{4}{3} \pi r_i^3 (\mu\text{m}^3) * \left[10^{-6} \left(\frac{\text{m}}{\mu\text{m}} \right) \right]^3 * 10^3 \left(\frac{\text{kg}}{\text{m}^3} \right) * 10^6 \left(\frac{\text{mg}}{\text{kg}} \right) \quad \text{Eq.1}$$

Which simplifies to:

$$M \left(\frac{\text{mg}}{\text{m}^3} \right) = \sum_i \frac{4}{3} \pi r_i^3 10^{-3} PNC_i \left(\frac{1}{\text{cm}^3} \right) * \left(\frac{\text{cm}^3}{\text{m}^3} \right) * (\mu\text{m}^3) * \left[\left(\frac{\text{m}}{\mu\text{m}} \right) \right]^3 * \left(\frac{\text{kg}}{\text{m}^3} \right) * \left(\frac{\text{mg}}{\text{kg}} \right) \quad \text{Eq.2}$$

Where:

M is the total particle mass

i is an impactor stage of the ELPI

r_i is the radius of the particles at impactor stage i

PNC_i is the measured particle number concentration at impactor stage i

The particle mass distribution is given in Figure 8, at the outlet of the water wash, with the BDU online (on) and by-passed (off). The total particle mass is estimated at 0.23 mg/m^3 with the BDU, and rises to 26 mg/m^3 when the BDU is by-passed. This a relatively low number (as mentioned, in aerosol emission regime, amine emissions are in the $100\text{--}1500 \text{ mg/Nm}^3$ range). Emission measurements of APBS-CDRMax, performed with an FTIR at sample port S3, indicate that by-passing the BDU did not influence the solvent emissions, see Figure 9. This seems to indicate that the particles are mainly composed of water, and an aerosol-based regime was not triggered.

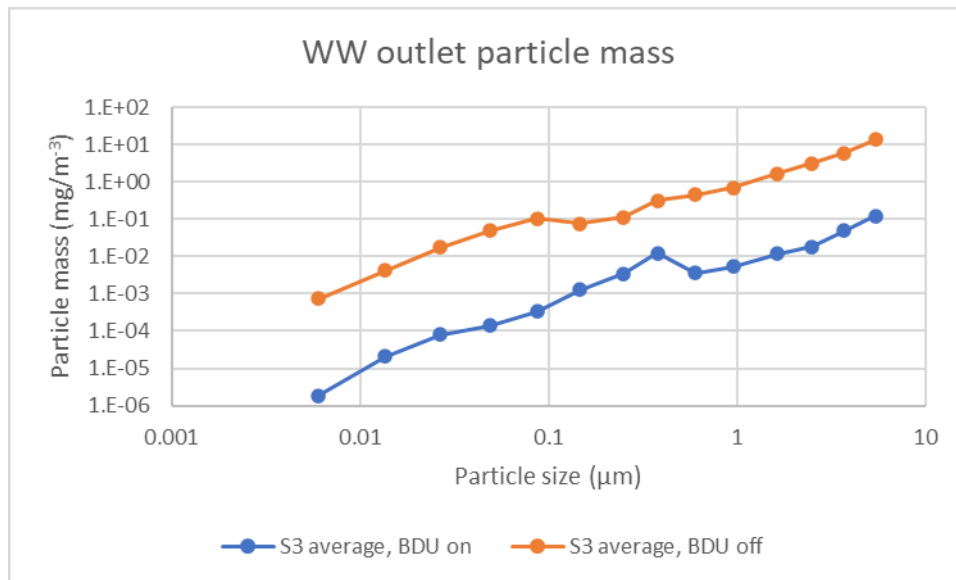


Figure 8 – Estimated particle mass distribution at the water wash outlet

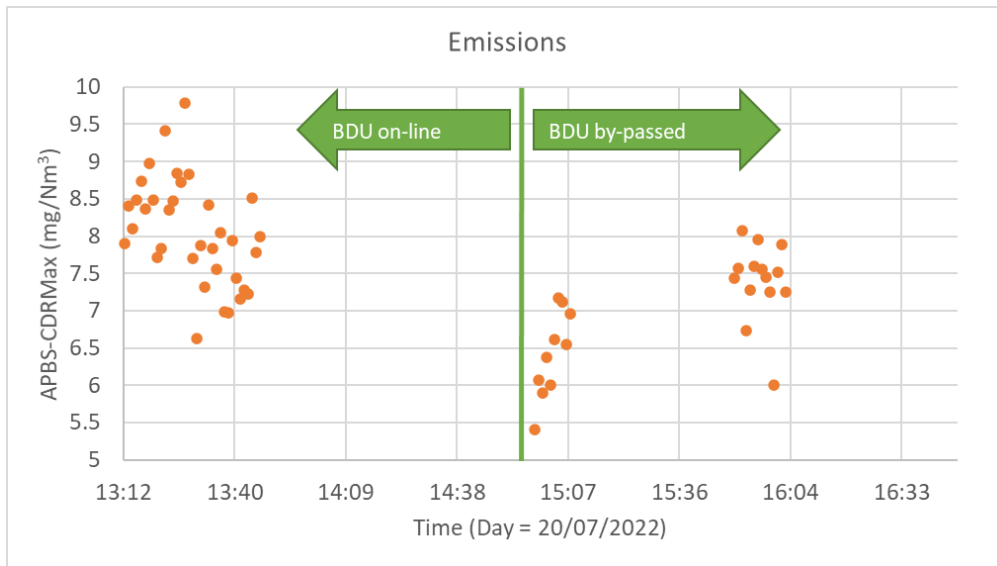


Figure 9 – APBS-CDRMax emissions measured with an FTIR at S3 (outlet of the water wash)

3.1 Comparison with previous campaign

The particle number distributions measured during the current campaign are compared to those of the previous campaign (March 2021) from Figure 10 to Figure 12. In March 2021, 3 measurements were performed at the BDU inlet (S1). The first two measurements (11:33 and 13:06) show less particles than measured in the current campaign. The third measurement (14:00), however, has higher particle numbers for particles above 0.02 micron (see Figure 10).

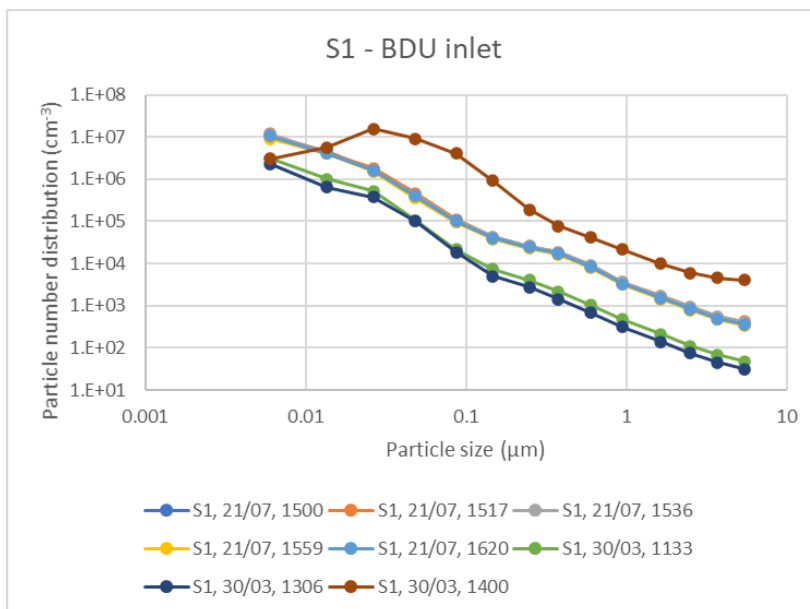


Figure 10 – Particle size distribution measured with the ELPI at sampling port 1 (BDU inlet), during the campaigns of March 2021 and July 2022

Two BDU outlet (S2) measurements were performed in March 2021, at 13:00 and 15:06. The first is in line with the measurements performed in July 2022 (see Figure 11), whereas the second measurement show higher particle count, particularly for particles below 1 micron. Two of the measurements performed in March 2021 (S1 at 14:00 and S2 at 15:06) show a different particle number distribution curve than all the other measurements, with a peak particle size around 0.02 micron. All the other measurements show a decay in particle numbers with increased size. It is postulated that this could be connected to differences in the upstream processes. All measurements performed at the water wash outlet with the BDU on (March 2021 and July 2022) are in good agreement.

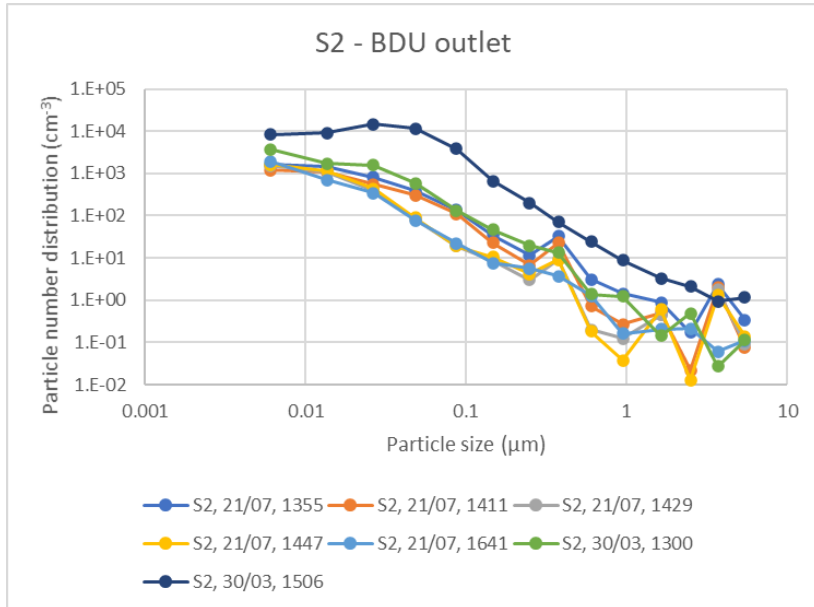


Figure 11 – Particle size distribution measured with the ELPI at sampling port 2 (BDU outlet), during the campaigns of March 2021 and July 2022

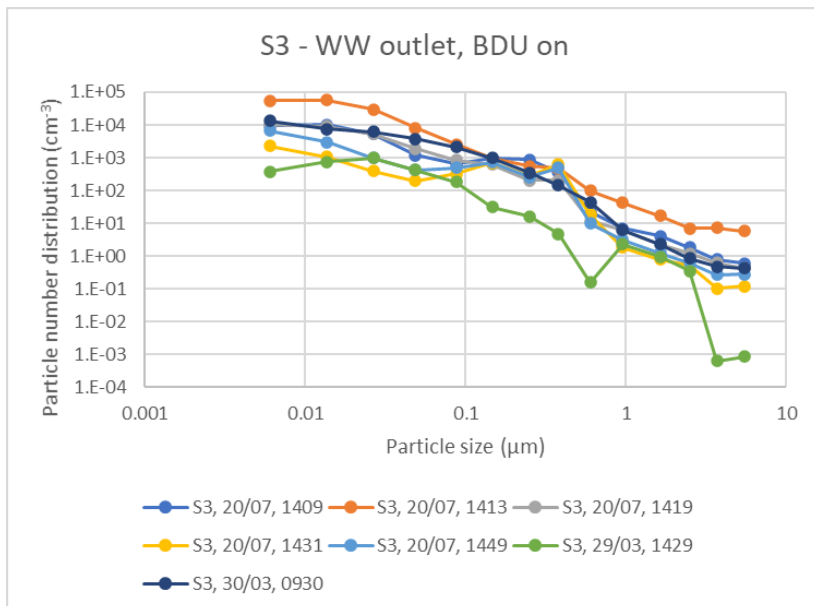


Figure 12 – Particle size distribution measured with the ELPI at sampling port 3 (water wash outlet), during the campaigns of March 2021 and July 2022

The particle numbers at the inlet of the BDU (given in Figure 10) were converted to mass using Eq.1. A total mass of 552 mg/m^3 is estimated based on the data measured at 14:00 on the 30th of March 2021. At that same time, the BDU was bypassed, and the emissions of MEA were registered by the FTIR located in port S3 (water wash outlet). The FTIR registered an average of 750 mg/Nm^3 of MEA being emitted, as shown in Figure 14.

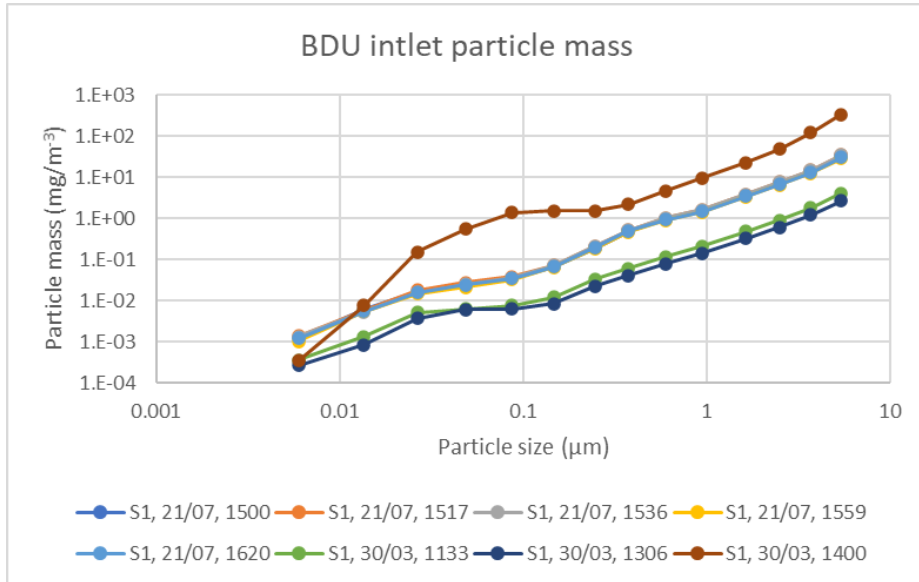


Figure 13 – Estimated particle mass distribution at the BDU inlet (S1) during the campaigns of March 2021 and July 2022

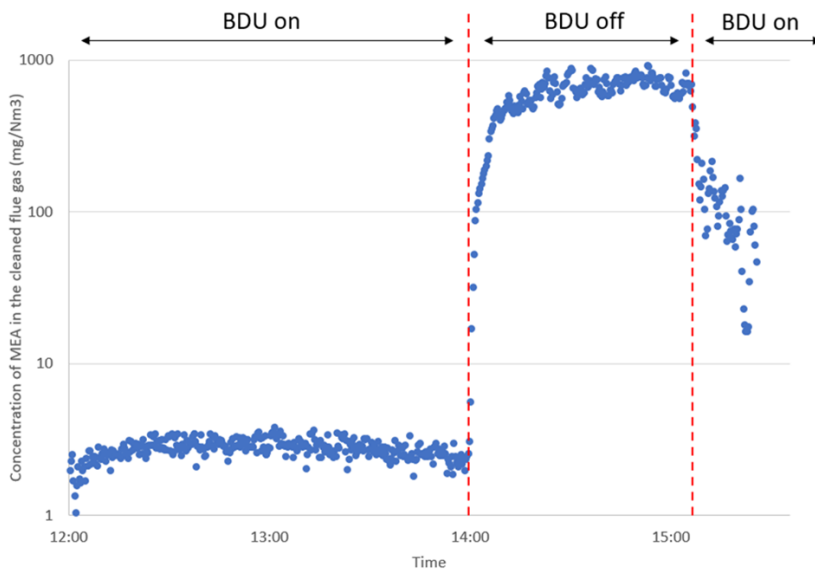


Figure 14 – MEA emissions determined by an FTIR at port S3 (water wash outlet). Source: [8]

3.2 Discussion on the BDU separation efficiency

The BDU separation efficiency (ε) can be calculated per impactor stage according to Eq.3 or for the entire range of the particle measurements, as per Eq. 4.

$$\varepsilon_i = 1 - \frac{PNC_{i,S2}}{PNC_{i,S1}} \quad \text{Eq.3}$$

$$\varepsilon = 1 - \frac{\sum_i PNC_{i,S2}}{\sum_i PNC_{i,S1}} \quad \text{Eq.4}$$

Ideally, when applying Eqs. 3 or 4, the measurements for the PNC at a given stage i should be known simultaneously for port S2 (BDU outlet) and port S1 (BDU inlet). However, in practice the ELPI is either measuring at port S1 or S2, so that these quantities are not determined simultaneously. There is the possibility that unknown upstream process disturbances change the PNC between one measurement and the other, which would lead to erroneous calculation of the separation efficiency. For the measurements performed in March 2021 and July 2022, the separation efficiency was performed by two methods. The *sequential method* considered the two sequential measurements of the inlet and outlet with the shortest time difference between them, in hopes of minimizing possible process disturbances. The *average method*, took all measurements done at that specific campaign into consideration, in hopes of lowering the effect of possible disturbances by averaging them out. Both methods give similar results for the present data. It is confirmed that the BDU is an efficient technique for lowering aerosol-based emissions, with separation efficiency above 99.8% measured in the campaign of March 2021, and even higher, 99.98%, in the campaign of July 2022.

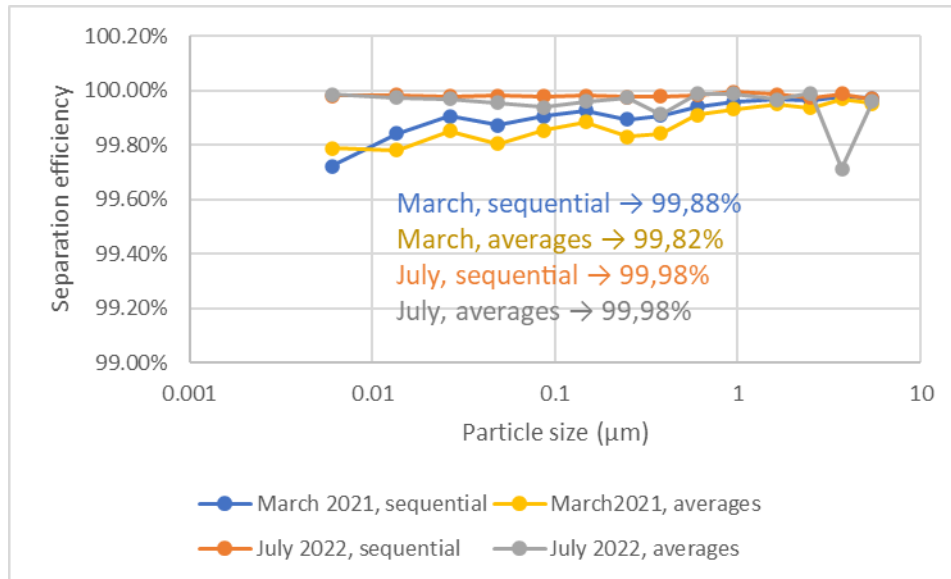


Figure 15 – Calculated BDU separation efficiencies per stage (Eq.3). The total efficiency (Eq.4) is given in the insert

The use of the BDU comes with associated costs. Capital costs are related to the initial investment on the equipment, while operational costs are due to extra energy consumption in the flue gas blower due to the pressure drop over the BDU, as well as costs associated with changing the candle filters over time. The average pressure drop measured at the Twence plant was 30 mbar on the 21st of July 2022, data in which the measurements around the BDU took place. In the ALIGN project, it was estimated that a BDU would have an OPEX of 0.09 € per 1000 m³ of flue gas, assuming a pressure drop of 20 mbar [5]. For 30 mbar, the OPEX would increase to 0.14 € per 1000 m³ of flue gas. Within

SCOPE, the cost estimates of emission mitigation technologies will be updated, based on the findings of the multiple test campaigns, to be performed in multiple plants.

Since its installation, the Twence BDU has accumulated ca. 10440 hours of operations. The separation performance in the 2022 campaign was higher than that in the 2021 campaign. Within the SCOPE project, tests will be performed on a regular basis to monitor the performance evolution.

4. Conclusions and Future Work

This work reports an experimental campaign measuring particles present in the Twence flue gas at the inlet and outlet of their BDU, as well as at the outlet of the absorber water wash, performed in July 2022. This campaign is compared to results from a previous campaign, performed in March 2021, right after the installation of the BDU on-site.

In the July 2022 campaign, the total mass of particles entering the BDU was of the order of magnitude 10^1 mg/m³. At this relatively low number, when the BDU was by-passed, no increase in the emissions were observed. On the other hand, the March 2021 campaign had a much higher mass of particles entering the BDU, with order of magnitude 10^3 mg/m³. When by-passing the BDU, an immediate increase in emissions was observed: from ca. 2.5 to 750 mg/Nm³.

It is confirmed that the BDU is an efficient technique for lowering aerosol-based emissions, with separation efficiency above 99.8% measured in the campaign of March 2021, and even higher, 99.98%, in the campaign of July 2022.

Within the SCOPE project, more measurement campaigns will be performed. The data obtained will be used to update separation models and cost correlations for the BDU, but also other emission control techniques.

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