Responsible dosing of iron(II)sulphate in a drinking water source; in control of coagulation.

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Abstract

In these proceedings we describe the work performed for the Dunea case during Physics with Industry 2016 at the Lorentz center in Leiden. The Dunea case centered around the dosing of iron(II)sulphate in a side branch of the river Maas, which is used as a source for drinking water. This dosing is done to remove phosphate from the river water, which prevents blooming of harmful blue algae. After an analysis of the problems of the current set-up, we give suggestions for better diagnostication and possible improvements.

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Introduction

Dunea supplies drinking water to 1.2 million people in the South Holland region in the Netherlands. One of the sources of the water production is from Managed Aquifer Recharge which is a process in which water is artificially resupplied or recharged in natural wells. Dunea uses a side branch of the river Meuse called the Afgedamde Maas as a source for recharging natural wells in the form of natural sand dunes in the Katwijk area. The water taken from the Afgedamde Maas needs to be treated before resupplying to the dunes. One reason for this is high content of nutrients from the agricultural area upstream from the sourcing site causing toxic blue algae bloom in summers. The blue algae grow particularly well in the presence of high concentrations of phosphate. This is dangerous because when blue algae decompose, they excrete cyanotoxins that are harmful to both humans and animals. Thanks to environmental regulation, both in domestic phosphorus pollution (from detergents etc.) as well as in agriculture, the phosphate levels in the Meuse has been declining steadily over the past few decades. Data supporting this trend is presented in Figure 1.



Figure 1: Left: blue algae bloom on Lake Crystal, Minnesota (USA). Right: decreasing concentration of phosphate in the Maas river from 1976 to 2014 in mg/L.

Still, it is necessary to reduce to amount of phosphate in the river water to prevent algae blooms. One common strategy is to use iron or aluminium to bind phosphates in the water⁵. In this direction Dunea has implemented a iron(II)-sulphate dosing system that has kept their reservoir free from blue algae blooms since 1997. The current implementation is due for an overhaul as the requirements have changed and the current pipe installation is reaching the end of its lifetime. Naturally, adding large amounts of chemicals can have adverse effects on the water quality and wildlife in the water so environmental concerns are important in the design. Running the plant at low efficiency is also costly in terms of chemicals and power consumption, further adding to the environmental footprint of the plant. Optimizing these factors is the main motivation for this project.

⁵ Parmar et al., ARPN Journal of Engineering and Applied Sciences, 2011



Figure 2: Schematic displays of the pipe that is currently used. a) Cross-sectional view: a small pipe for the dosing of iron sulphate with a diameter of 28 mm is embedded in a bigger pipe for the air with a diameter of 110 mm. Both pipes have nozzles protruding from the top. b) Top view of one group of nozzles. Every group of nozzles contains three air nozzles with a diameter of 2,4 mm and one smaller iron sulphate nozzle with a diameter of 1.5 mm. c) The total pipe is about 100 meters long and contains 70 of such groups of nozzles.

Current implementation and problems

The current installation consists of a pipe, approximately 100 m long along the river bottom, a cross section can be seen in Figure 2 a). The pipe carries air as well as liquid phase iron(II)-sulphate dose in a smaller pipe inside. A schematic image of the details of the the nozzles and the distribution of the nozzles are shown in Figure 2 b) and c) respectively. The purpose of the air is twofold, i) aerating the river to compensate for the oxygen consumption from the reaction

 $4 F e^{2+} + H_3 O^+ + O_2 \Rightarrow 4 F e^{3+} + 6 H_2 O$

and ii) enhancing the mixing of the constituents.

One of the issues the current system has is that the air nozzles tend to clog a few times every year, which has associated downtime and maintenance. The operators notice the clogging of the nozzles by an increase in the pressure of the air and iron sulphate pipes. The pressure profiles in both pipes are displayed in Figure 3. For the air pipe the data is unambiguous: the pressure slowly increases over time until the situation becomes unmanageable and a diver is hired to clean the air nozzles. Cleaning of the nozzles leads to a drop of the pressure; one such cleaning in June of 2016 can be seen in Figure 3.

However, the pressure profile in the iron sulphate pipe seems to show a global decreasing behavior from March until July. This could be a consequence of the fact that the viscosity of most solutions decreases as their temperature increases, something which happens exactly in these months. We estimated the associated pressure drop and it seems to be in the right value range based on the data depicted in Figure 3. However, this is an approximation and at this point it is difficult to draw any conclusions about the iron-dosing pipes and if, when and

why they clog. With the data at hand it seems it is mostly the air pipes that clog so the focus of our efforts will be on the clogging of the air nozzles.



Figure 3: Left: the pressure (kPa) in the air (orange) and iron sulphate (blue) pipes from January 2016 to November 2016. The sudden drops to zero correspond to the switching off of the pumps. Right: zoom in on the data of the air pump. The pressure increases to a maximum value around the beginning of June, when a diver unclogged the nozzles. After that the pressure starts to increase again.

An additional issue is that the system uses a lot of energy, particularly the air pumps consume 21 kW. All iron sulfate dissolves in water to give the same aquo complex $[Fe(H_2O)_6]^{2+}$. When mixed with air, iron (II) sulphate solution further gives iron(III) sulphate and iron oxide according to the reaction:

12 $FeSO_4 + 3 O_2 \rightarrow 4 Fe_2(SO_4)_3 + 2 Fe_2O_3$

Iron (III) sulphate further gives iron (III) charged molecules to form Iron hydroxides

Fe(OH)₃ with water: $3Fe_2(SO_4)_3 \rightarrow 2Fe^{3+} + 9(SO_4)^{-1}$

Ferric hydrate (FeSO₄.7H₂O) contains 55 g per mole of iron. If it is assumed that all iron from the iron sulphate solution oxidizes, 64 g of oxygen molecules is required to convert 56 g of iron to form iron phosphate FePO₄. As oxygen is one fifth of air, 320 g of air is required for 56 g of iron to Fe³⁺ and further to iron phosphate. If we consider to dose, 25 tonnes of iron sulphate solution of which 5 tonnes is iron, it would require 30 tonnes of air. The density of air (at atmospheric conditions) is 1.205 kg/m³. The maximum flow rate at which pump can inject air is 0.2 m³/s, so the pump can inject air at 0.245 kg/s. Therefore to inject 30 ton of air at a maximum power of the pump, it will take ~35 hours, assuming all air is mixed with the iron.

To summarize the objectives of the project we want to lower the

- Chemical dosage;
- Downtime;
- Maintenance costs;
- Power consumption.

Considerations

The objective of the water treatment is to keep phosphate levels low enough to keep the water clear and divest the water of nutrients to avoid blue algae blooms that could be detrimental for water production. Many considerations have to be taken into account in this problem as the complexity is very high.

The Afgedamde Maas is a dead river arm of the Maas. The intake is 6km from the entrance, the dosing facility is 4 kilometers from the intake. Hence there is much space for the reactions for the water treatment. The net flow is 10 meters per day and is caused by the pump at the end of the river. The main waterflow is caused by the tides. This tidal wave moves the water 2.5 kilometers in 6 hours. This flow provides significant passive mixing power. There are two distinct passive mixing processes: vertical and lateral mixing. For both processes we can associate a specific length scale depending on the dimensions of the river, and a time scale depending on the flow speed. Passive vertical mixing in the river occurs on length scales of around 70 meter, which takes up to 6 hours. This time is approximated to be longer than the active period of the iron-salt mixture, hence active mixing is necessary to ensure sufficient mixing. Careful chemical analysis of the dynamics of the reactions involved is necessary to make any confident conclusions.



Figure 4: Liquid film coefficient K_L in cm/hr as a function of the diameter d_B of the air bubbles coming from a air nozzle. Optimal aeration of the water, corresponding to the highest value of K_L , occurs at a bubble diameter of about 2,5 mm. Empirically, this corresponds to typical nozzle diameters of 0.1 to 0.3 mm.

As mentioned earlier the injected air is responsible for mixing and for aeration, or oxygenation of the river. The size of the air bubbles is critical for the diffusion of the air into the water. This is a way to control how efficient the aeration is in terms of the amount of gas that is actually absorbed in the water, as can be seen in Figure 4. A large part of the expected savings are associated with costly operation of the aeration pumps. Stoichiometrically optimizing the aeration to obtain a minimum oxygen level for which the desired reactions will still occur sufficiently is probably crucial to minimize air pumping. This will in turn give a boundary condition for the minimum pumping, with a given bubble size. Finally this amount of pumping still has to be enough to mix the solutions sufficiently.

The iron is put in the river in the following way. Dunea dissolves commercially available $FeSO_4$ in water in tanks on the river bank in batches of 25 ton of salt. This yields Fe^{2+} and SO_4^{2-} ions. When this fluid is injected into the water stream, the Fe^{2+} reacts with O_2 to form Fe^{3+} . The iron oxidation is a rather slow process with a typical time between 1 and 200 minutes. Only the Fe^{3+} ions can react with PO_4^{3-} to form solid $FePO_4$. Besides the phosphate capture, Fe^{3+} also reacts with OH^- . This process is very fast, and hence a more than stoichiometric ratio should be added in order to have phosphate removal. Such iron-hydroxides have the beneficial properties that they form flocks, who coagulates and removes sediments from the stream.



Figure 5: Three mixing conditions of Fe^{3+} in the river.

The phosphate removal process strongly depends on both the dosing and the mixing properties. There are three regimes, as indicated in Figure 5. When the Fe³⁺ concentration is too low, no phosphate is removed, only sediments are removed. On the other hand, when the iron solution is not mixed, the Fe³⁺ concentration is very high and hence the phosphate removal is good, but only in a small volume. The optimal mixing strategy is thus dependent on the dose and dosing speed. More numerical modelling is advised in order to optimize the total injected dose.

Additionally, there are many considerations that are specific for the conditions of the dosing station, everything from biomass content, flocculant size and biological activity to take into account. In general, in winter algae are not growing and hence the dosing can be much lower, hence the dosing of iron is only 20% of the summer time dosing, as shown in Figure 6.

During the oxidation of Fe^{2+} to Fe^{3+} oxygen is removed from the water. This oxygen loss has to be compensated, which is currently implemented by bubbling compressed air through the water. This supply of air also helps with mixing. Currently, the air supply is kept constant even though the iron dosing is only a fraction in winter compared to summer. Adjusting the air supply to the dosing presents a large potential saving.

Due to the aforementioned complexity of these systems and the data on the actual pumping system available, most of these questions cannot be answered quantitatively from theoretical approaches in physics alone. One can attempt engineering approximations, which is what has been done in this workshop. Another promising approach would be to attempt looking at these problems with chemistry method and analysis.



Figure 6: Planned dosing of Iron sulphate (kg) in the different weeks of 2016. The amount ranges from less than 5000 kg in winter to 25000 kg in the summer. In the summer there is more biological activity due to a higher average temperature of the water, resulting in a more pressing need to remove the phosphate from the river water to prevent (blue) algae blooms.

Approaches

Disentangling all the variables and processes is a daunting task. An alternative approach to this would be to simply measure the phosphate levels for different mixing and dosing methods and pressures and ensure that the levels are acceptable. With this information in hand, more substantial claims can be made. Nevertheless some general advice can be given.

We will first optimize the geometric design of the set-up, such that it is expected to clog less frequently, or ideally not even at all. Depending on what the clogging mechanism is, a backflow-preventing valve may need to be installed. These may be simple mechanical valves or electronically operated valves. Despite all these measures, the nozzles may still clog after a longer period of time. Therefore, we have also considered active cleaning options which automatically unclog the nozzles. In the following all approaches will be described in some detail.

Geometric optimization of the pipes and nozzles

From an engineering point of view, a geometric optimization of the pipe and nozzle dimensions seems to be the most natural candidate. Can we think of simple changes we can make to the pipes and nozzles, to prevent them from clogging as quickly as they do now?

The piping system is close to the end of its lifetime and needs to be replaced in the next few years. Naturally, the system upgrade is an opportunity to also optimize the geometry of the piping and the distribution of the nozzles. The hydrodynamic system is governed by a rather simple equation:

 $\Delta P = \alpha LQ_{T}^{2}/(d^{5}N^{2}),$

where ΔP is the pressure difference across the nozzles, L is the length of each nozzle, d is its diameter, Q_T is the total flow through all the nozzles, N is the number of nozzles and α is a constant related to the properties of the flow.

Based on this relation, there are a few parameters that can be tuned to get optimal conditions: the length L, the diameter d and the number of nozzles N. We also need to take into account a few constraints and, naturally, the goals of the optimization:

- To reduce dosing, we would like to **reduce the minimum flow** that the system can handle (Q_{min}). Currently this value is: Q_{min} = 200 l/h and our goal is to reduce this at least by a factor of 2.
- The maximum flow the system should be able to produce is fixed and has to do with emergencies, i.e. outbreaks of blue algae. The maximum flow rate should be Q_{max} = 1000 l/h.
- To prevent clogging and/or reduce clogging maintenance costs, a combination of **reducing the number of nozzles** and **increasing the nozzle diameter** would be a possible direction.
- The minimum operating **pressure of the pump** should be kept well **above 0.6 bar** to counteract the pressure of the water in the river at the depth of the pipe and prevent water from entering the pipes.

To find the optimal solution satisfying the constraints, we ran calculations varying each of the proposed parameters. In Figure 7 we summarize the results for both the $FeSO_4$ and the air nozzles.



Figure 7: A plot of the required pressure ranges for the pumps as a function of nozzle length and diameter with the goal of extending the dynamic range of the flow. Left: dynamic range for the $FeSO_4$ pump for a flow range of 70-600 l/h. Right: dynamic range for the air pump for a flow range of 5000 - 9000 l/m. The white space signifies that a solution could not be found for the given set of parameters and constraints.

As mentioned above, it is of interest to increase the diameter of the nozzle, assuming that a larger diameter would lead to less clogging. Increasing the length of each nozzle, should pose no issue in general. However, installing very long nozzles may yield them more brittle and they may break when subjected to stronger turbulence. It is thus important that the nozzles are kept reasonably short (< 40 cm) and as wide as possible, provided that they can supply the minimal required flow with the minimum pressure difference that the pump can handle (in this case, it is assumed that the pump can have a pressure resolution of at least 0.01 bar). From Figure 7 (left panel) we can conclude that an $FeSO_4$ nozzle with a length of 30 cm and a diameter of 1.9 mm would be the optimal given the constraints. Using pressures between 0.6 and 2 bar, one should be able to cover the entire flow rate range for the standard operating conditions (70 - 600 l/h). Doing this, the minimum possible flow would be reduced by a factor of 2, which would prevent overdosing in periods of low tide and would eliminate the need of diluting the iron sulfate solution in the winter season.

A similar calculation for the air nozzles is shown in Figure 7 (right). In this case, however, there is an additional consideration that we need to take into account. The efficiency of the aeration depends strongly on the air bubble size, which in turn depends on the nozzle diameter. The ideal bubble size for aeration according to literature is around 2.5 mm, which would require nozzles of around 1mm in diameter. This is, of course, not optimal for the system taking into account all the aforementioned considerations. There is a trade-off between increasing the diameter to get less clogging and decreasing it to get smaller bubbles. Closest to the optimal value would be a diameter of 4.2 mm which would require a very short nozzle (5 cm). It is important to note that such a configuration should be able to handle much smaller flow rates (5000 l/h, compared to the standard 8000-9000 l/h) which, in periods of low dosage, could significantly cut down on power consumption for pumping.

Mechanical valves

In case of an emergency, such as a pump failure or a power cut, the pressure in the iron dosing pipe and air pipe may decrease to levels below 0.6 bar. This will allow river water to enter the nozzles and pipes. This is undesired because river water in the air pipe will be difficult to remove, and river water in the iron dosing nozzles may lead to initiation of the precipitation mechanisms, and consequent clogging of the nozzles.

An obvious way to prevent backflow of river water into the system is to use backflow-preventing valves. In industrial settings such valves, called *check valves*, are often installed in pipe systems. However, we have a more specific need here: we need a valve which automatically closes the *exit* of the nozzles as soon as the flow (either iron solution or air) is stopped or in danger of reversing. This could theoretically be accomplished by a ball and spring system as depicted in Figure 8 below. Under normal circumstances, the flow should be strong enough to overcome the spring force.



Figure 8. Examples of mechanical backflow-preventing valves which close the exit of the nozzles as soon as the flow through the nozzle stops or is in danger of reversing. Under normal operation, the flow should be strong enough to overcome the spring force.

A back-of-the-envelope calculation can be made for the required spring constants. Assuming that all momentum present in the fluid flow through a nozzle is just stopped by the spring at a spring extension of the order of a few mm (the nozzle diameter), we find that a typical flow force $F_{flow} = \frac{1}{2}\rho v^2 A$, with A the nozzle cross-sectional area, should be balanced by a spring force $F_{spring} = kx$, with k the spring constant and x the required spring extension, which we take equal to the nozzle radius. For the air nozzles the flow is relatively strong (with air velocities of the order of 10^2 m/s), which leads to a spring constant of the order of 10^2 N/m. However, the flow velocity in the iron dosing nozzles is so low (of the order of 1 m/s) that the spring constant for these nozzles would need to be of the order of 10^{-2} N/m. This is probably so weak that any (temporary) turbulent flow in the river can also open such a valve. This is a serious problem, because clogging is likely occur when river water can enter the iron dosing nozzles.

In summary, the air nozzles may be automatically closed by installing mechanical valves, but the iron dosing nozzles need a valve which does not depend on the momentum (or pressure) of the iron solution at the nozzle exit.



Figure 9: Proposed wiring schemes for electrically controlled nozzle valves. Top: Separate groups of nozzles for the center of the river bed and the sides. Bottom: alternate (odd/even) configuration useful for intermittent dosing. Each color represents a group of nozzles.

Electronic valves

One approach to overcome this issue would be to install an electronic valve for each nozzle. This has the obvious advantage that everything could be electrically controlled, so during maintenance, power outages and even extremely low dosing, the valves could be closed remotely, preventing backflow and cutting down the dosing completely. Such valves could also reduce the chance of clogging by opening and closing more frequently. Another step forward could be grouping the nozzles (i.e. valves) in two or more separate groups that can be individually controlled. In such a way one could envision control schemes where the nozzles on the sides of the river bed are more frequently opened than the ones in the center to compensate for the lower dosing on the river sides (see Figure 9, top panel) or even a scheme where even and odd nozzles are grouped separately, to provide for the option to halve the flow in period of low dosing (Figure 9, bottom panel). The installation of electronic valves would also enable implementing the intermittent dosing scheme more easily, as electrical pulses could be applied to the nozzles such they are open only a certain percentage of the time. The major

disadvantage would be the installation cost of such a system and the fact that there may be sensitive electronics under water.

Active clogging prevention

Several different approaches were suggested, ranging from temperature induced expansion of the nozzles and piezo-induced vibration, to ultrasound dissolving of the clogs. However, all of these seem to be unfeasible in the long term. One has to take account of the fact that the pipe should be situated in the river for a rather long time. High tech solutions like piezo-induced vibrations, or even local heating of the nozzles therefore seem to be far-fetched for the current stage of the system.

Conclusions

Below we list some substantial advice on what questions should be answered and some nozzle design and dimensions. The first two advice are meant to obtain a better characterization of the system. The rest are are on options to optimize the system:

- 1) One important question to answer is what the nature of the substance is that clogs the air nozzles. On-site human observations describe it ranging from a "fluffy" to a "hard" substance. X-ray diffraction at several sites with similar clogging problems have resulted in the identification of calcium sulfate and calcium phosphate as being the clogging agent.⁶ Other data indicates it is mainly ferrihydrite.⁷ Due to this uncertainty, it is desirable to harvest some of the clogging material and investigate what it is made of.
- 2) More systematic measurements have to be done to characterize the system better. At the very least the phosphate, oxygen and iron concentration before and after the dosing station should be measured at different dosing conditions. It is very possible that the air dosing can be halved in winter time but no claims can be made without knowing the phosphor level responds to this. In depth knowledge about the chemistry and mixing is required.
- 3) At the moment the air pump has only one power option. Using an air pump that has different power levels will allow a reduction of the power level in the winter period. In this period less iron sulfate is dosed and consequently less oxygen is removed from the water. Therefore, a pump with a lower power suffices. This will reduce the energy costs in the winter period considerably because the necessary power scales with the volumetric rate to the power of 3.

⁶ J.A. Müller, W.C. Boyle, and H.J. Pöpel, "*Water Quality Management Library (Volume 11). Aeration: principles and practices*", CRC Press (2002).

⁷Medina et al., "*Iron-hydroxide clogging of public supply wells receiving artificial recharge*", Hydrogeology Journal (2013).

4) If significant improvements in the dosing are to be obtained, it would be advisable to investigate the possibility to use a control system (feedback or feed forward) investigated. This is common practice to implement in wastewater plants and should in principle be possible to implement here too^{8,9}. One difficulty with the control system route is to find a feasible input parameter to feedback on. Just using phosphate levels is insufficient as the phosphor level goes down when there is an algae bloom in the river. This means that during a algae bloom a feedback system would lower the dosage instead of increasing it. However, if one can model the algae risk well or find a suitable marker or combination of markers that one can measure, it should be feasible. In 2016 Dunea commissioned a report that produced a model for the phosphate levels and we see that it is quite accurate but it is indeed less accurate in summer. The possible savings in phosphate levels can be approximated by comparing the dosing levels in Figure 6 with the actual measured values of phosphor in Figure 10. As there is a large discrepancy between the two, there should be considerable savings possible. Alternatively, measurement of the phosphor level could be done in the river Maas and use this as the feedback reference. Because of its higher flow velocities it is much less likely to develop algae blooms and it therefore will not have the same issue with misleading phosphor levels.



Figure 10: Figure from a commissioned report from the Water Research Institute in 2016. Using a simple model they make predictions on the phosphate level in blue to be compared with the measured values in red. The dashed blue line is the actual dosing and the dashed green line is the measured phosphor level in the Maas. If data like this is available on at least monthly basis, feedback should be feasible.

⁸ <u>http://www.worldpumps.com/view/316/control-of-chemical-dosing-in-wastewater-treatment/ Retrieved on</u> <u>Nov 24, 2016</u>

https://www.epa.ie/pubs/advice/drinkingwater/Drinking%20Water%20Advice%20Note%2015%20WEB.pdf

5) At the moment the nozzles are made of arnite (a high strength and rigid plastic). Clogging of nozzles may be enhanced by a sufficiently high strength of adhesion between sediments / precipitates and the nozzle material. It is advised to investigate the performance of different nozzle materials (or coatings) with relatively low strength of adhesion, such as PTFE (teflon) or ceramics. To optimize this, the clogging agent needs to be analyzed as per point 1.



Figure 11: Different pipe designs. a) Original pipe design. b) Pipe design in which the air nozzle (grey) is longer than the iron sulphate nozzle (yellow). c) Similar to b). d) Rotated design in which the air nozzle pipe is situated below the iron sulphate nozzle, hence propelling the iron upwards and preventing clogging of the air nozzle.

6) Replacing the current piping with new dimensions. Figure 12 shows the missing nozzles in the current system. A missing nozzle gives poor performance down the pipe from a clogged nozzle, as would be expected from a large pressure drop as a result from a missing nozzle. A new design for a nozzle can be seen in Figure 11. The angle on the air pipe is meant to eliminate backflow from water into the air pipe when the pump is turned off. Further, if possible a check valve should be implemented, either on the main pipe to keep backflow from occurring or (if feasible with the choice of pressure range) on the nozzle itself.

_1	Н	26	BD	50	Н	OP=Open
_2	OP	27	BD	51	D	
3	Н	28	BD	52	D	N=Nozzel Gone
_4	BD	29	OP	53	OP	
_5	Н	30	N	54	D	H= Half open
6	Н	31	BD	55	D	
_7	BD	32	BD	56	D	BD= Almost closed
8	BD	33	BD	57	D	
9	Н	34	BD	58	D	D=Closed
10	BD	35	OP	59	D	
11	D	36	D	60	BD	
12	Н	37	BD	61	D	
13	Н	38	BD	62	D	
14	OP	39	D	63	N	
15	OP	40	D	64	BD	
16	BD	41	OP	65	BD	
17	OP	42	Н	66	?	
18	OP	43	OP	67	?	
19	OP	44	BD	68	?	
20	OP	45	D	69	?	
21	OP	46	D	70	?	
22	OP	47	OP			-
23	OP	48	OP			
24	Н	49	Н			
25	Н	50	Н			

Figure 12: Measurements on the clogging of the different groups of nozzles, numbered from one side of the bank (1) to the other bank (70). The measurements are done by visual inspection over the pipes by boat and approximating the amount of air bubbles coming from each nozzle. A striking feature is the fact that the first thirty nozzles are performing relatively well, whereas the nozzles after that are clogged much more. This is due to the fact that the nozzle groups 30 and 63 are gone completely. This yields a big pressure drop after these nozzle groups, resulting in more clogging in the following nozzles.