Solute Concentrations Vastly Exceeding "Ultrapure" in Thereof Dependent Detergent Free Laundry Due to Practical Limits of Dilution and Concomitant Release of Solubles From Textiles *per se* – A Case Study of a Detergent Free Washing Machine of the Landlord MKB's (Malmö, Sweden)

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Abstract

The solute levels (Total Dissolved Solids, "TDS") of the outgoing washing water following each washing cycle of a typical detergent free washing machine (MKB Fastighets AB, "Rönnen", Malmö, Sweden), which efficiency relies on the claimed inherent high cleaning power of "ultrapure" water, were estimated by electrical conductivity measurements. A set of typical personal laundry (3,97 kg) was washed on December 12, 2019 using a program labelled "for heavily soiled laundry" (Sv: "För hårt smutsad tvätt") employing a feed water with relatively high purity (TDS = 0,4 mg/L). The TDSvalues of the outgoing water of the associated five washing cycles were determined to be 351, 247, 42,7, 21,7, and 16 mg/L, respectively. A developed predictive model, being highly consistent with an apparent dilution factor (D) of 3,32 and a constant solute contribution (C_L) from the textiles per se of 8,3 mg/L, was used to simulate the employment of a ten times less pure feed water (TDS = 4,0 mg/L) under otherwise identical or slightly modified conditions. It was thereby found that close to an identical TDS-profile could be achieved with such a cheaper and more environmentally friendly feed water. In conclusion, the results of this study indicate that the laundry was never, in any practically significant meaning during any cycle of the washing procedure, subjected to washing water with a purity which generally is to be considered as "ultrapure". Such an incongruity, between the experimental results of this study and the current claim of extraordinary cleaning properties of "ultrapure" water or the like, is clearly warranting further and extended open research in this area. It is suggested and hypothesized by the author that any subjectively determined acceptable cleaning results, in this and other similar detergent free washing machines, may be explained by a combination of user bias and the employment of mechanically and rinse-wise optimized washing programs, rather than to any hitherto unknown inherent property of water.



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Background

Textiles, typically personal clothes, towels and linens, are traditionally cleaned in laundry processes by employment of various kinds of detergents collaborating with common tap-water, e.g. in laundry machines which provide the additional essential mechanical rubbing and agitation. The employment of e.g. a liquid or powdered laundry detergent in such a process is motivated by the established concept of "detergency", comprising roll-up-, emulsification- and solubilization-interactions between surface active molecules and non-polar matter, e.g. greasy soil, to be separated from the textiles.¹

The necessity of detergents in laundry has, as can be noted from various media reports,² recently been challenged by intercessors of detergent free laundry systems. One type of such a system, currently marketed as having the benefits of being energy saving and enabling an environmentally benign way of doing laundry, comprises a water purification unit feeding purified input water, typically called "ultrapure water" or "DIRO[®]-water", to a customized washing machine of standard type. Spearheaded by the Swedish company Scandinavian Water Technology (SWATAB),³ various awards⁴ and substantial Swedish governmental economic sponsoring⁵ have been granted in support thereof. As a theoretical basis for the supposedly equivalent or superior efficiency of "ultrapure water" or "DIRO[®]-water", in comparison to the corresponding detergent aided laundry, it is claimed that such a highly pure water has intrinsic dissolving and/or suspending properties that differs significantly from a less pure water, e.g. tap-water.⁶ The following hypothesis may be formulated on basis of such a claim:

"Water alone, but with a high degree of purity, has at least the same ability to dissolve or suspend non-polar matter as common water in combination with one or several detergents, under otherwise identical conditions."

Early reports on unexpected relatively stable surfactant free emulsions in deionized and de-gassed water,⁷ provided some support to above hypothesis. However, in all of these cases the water concerned was de-gassed by several freeze-pump-thaw cycles and a more recent report suggests bicarbonate, generated by the trapping off external carbon dioxide, to be essential for the stability of the formed emulsions.⁸ Hitherto, all additional evidence in favor for or against above hypothesis consist of anecdotal observations lacking scientific stringency and internal experimental reports.⁹ In addition, a scientifically serviceable suggestion on the cleansing mechanism-of-action of water with a high degree of purity in laundry or other applications, is yet to appear.

The present study was conducted in order to gain objective data of use for e.g. the planning of future studies in this area and as a factual basis for scientific discussions and debate. Hence, the purity of the water in an operational detergent free washing machine of the type explained above, serving a multitude of tenants in a residential area in Malmö (Sweden), was monitored throughout the laundry program. To the best of the author's knowledge, this is the first time such data is being reported. Previous data from the patent literature¹⁰ on what purity threshold differs a "pure water", with claimed inherent unexpected but extraordinary washing properties, from a "common water" such as e.g. tap-water, without such properties, may thus be compared with real case data from an operational washing machine with alleged acceptable cleaning efficiency.

Experimental

The study and the data collection were conducted at the municipally governed landlord MKB Fastighets AB's internal laundry facility for student tenants at the residential area "Rönnen" in the city of Malmö, Sweden, on December 12, 2019. Representatives of said landlord and of SWATAB,³ were at all times present during the study to monitor the execution of the study and the data collection procedure. Of the two available identical washing machines of front-loading type (Electrolux), the right-hand side one was used. At the point in time of this study, the tenants had been using them as their main facility for washing their personal clothing and similar textiles, in line with the instructions that no washing powder or detergents are to be used, and that further heating of the washing water above ambient temperature is not necessary for a satisfactory washing result.

The level of Totally Dissolved Solids (TDS) in all water samples were estimated by measurement of the thereto relating electrical conductivity using a hand-held conductivity meter (Greisinger, GMH 3431) in its factory settings, calibrated with a 1413 uS/cm calibrating solution according to the operating manual. All conductivity- and TDS-values are reported as measured by the instrument being set to operating in "non-linear temperature compensation for natural waters" ("nLF") in accordance with EN 27888 (Table 1 and Table 2). The corresponding actual temperature of the sampled water in any such measurement is also reported.

The conductivity/TDS of the local tap water at the site of the study (Malmö, entry 1), of the feedwater of the washing machine (DIRO[®]-water, entry 2), of the milli-Q[®]-water used for rinsing equipment and buckets (entry 3), as well as the weight of the dry laundry before washing (entry 4) and the moist ditto (entry 5) after the complete finishing of all washing programs, i.e. a first wash followed by a second wash, is collected in Table 1. The results of a separate experiment conducted in Lund on a relatively small reverse osmosis unit (Aqua Medic, Easy Line 90) for the desalination of tab water, are also collected in Table 1. Depending on the permeate/concentrate-ratio (EF), a varying degree (entry 8-10) of desalination of the input water (entry 7) was thereby observed.

A collection of used and mildly soiled personal clothing comprising underwear, socks, T-shirts and towels (totally 3,97 kg, Table 1, entry 4) were washed twice, in a first wash directly followed by a second wash without removal from the machine and, in both cases, employing a program labeled "for heavily soiled laundry". The moist laundry after finishing of the second and final wash (totally 5,42 kg, Table 1, entry 5) comprised 26,8 % residual moisture after the final centrifugation thereof. Each wash consisted, as observed, of five trailing washing cycles consisting of the following steps (i)-(iii): (i) inlet of feed water enough to enable washing of the laundry; (ii) washing by repetitive spinning of the central washing drum clockwise and counter-clockwise; and (iii) output of used washing water via a large external hose leading to an open draining gutter. The output used washing water was collected in pre-rinsed (milli-Q[®] water) 14 L plastic buckets for immediate subsequent withdrawal of three separate 40-50 ml aliquots (50 ml PP-tubes, Sarstedt AG,nr. 62.547.254) for analysis and storage for potential future analyses. With exception of the output water from the first cycle, which was significantly less in volume as compared to the output water from subsequent cycles, only about 75 % (visually estimated) of the output water was collected in the respective bucket due to this having a volume falling short of the typical output volume. The conductivity/TDS and additional data relating to each washing cycle (flush #, "n") of the first and second wash (wash #, "m") is depicted in Table 2.

Object	Entry #	Conductivity (uS/cm)	Cond.Temp (°C)	TDS (mg/L)	TDSTemp (°C)	Weight (kg)	Rate (g/min)
Tap-water (laundry) (Malmö)	1	338	12	335	12,1	N.A.	N.A.
DIRO-water (laundry)	2	0,4	17,2	0,4	17,2	N.A.	N.A.
cleaning-water (milliQ)	3	1,1	7,4	1	7,4	N.A.	N.A.
laundry pre-washing	4	N.A.	N.A.	N.A.	N.A.	3,97	N.A.
laundry post-washing wet	5	N.A.	N.A.	N.A.	N.A.	5,42	N.A.
laundry post-washing remaining water	9	N.A.	N.A.	N.A.	N.A.	1,45	N.A.
DO-water - input water (Lund)	7	181,7	12,2	181,8	12,3	N.A.	N.A.
DO-water - output @ EF=2,39	8	6,3	15,9	6,2	15,8	N.A.	N.D.
DO-water - output @ EF=5,07	6	4,2	15,6	4,2	15,7	N.A.	N.D.
DO-water - output @ EF=16,46	10	3,1	15,1	3,1	15,1	N.A.	85,3

<u>Table 1</u>: A collection of the results of various conductivity/TDS-, weight- and permeate/concentrateratio- (EF) measurements.

flush#"n"	wash # "m"	Conductivity (uS/cm)	CondMesTemp (°C)	TDS (mg/L)	TDSMesTemp (°C)	weight (kg)
1		352	20,1	351	20,6	7,58
2		245	20,2	247	19,6	>13,07
ſ	1	43,8	18,5	42,7	20,4	>13,05
4		21,7	21	21,7	20,2	>12,86
S		16,2	19,2	16	20,1	N.D.
9		N.D.	N.D.	N.D.	N.D.	N.D.
7		N.D.	N.D.	N.D.	N.D.	N.D.
8		N.D.	N.D.	N.D.	N.D.	N.D.
9	~	N.D.	N.D.	N.D.	N.D.	N.D.
10 (average of A-C below)	ı	8,8	19,7	8,7	19,2	N.D.
10A		8,9	19,4	8,7	18,4	N.D.
10B		8,4	19,6	8,3	20,3	N.D.
10C		9,1	20	9,1	19	N.D.

<u>Table 2:</u> Conductivity, TDS and additional data relating to each washing cycle (flush #, "n") of the first and second wash (wash #, "m").

Discussion

The laundry washing cycles can be regarded as essentially being a series of dilutions of solutes from previous washing cycles dissolved in the remaining water of the foregoing moist textiles, in parallel with solubilization of materials being physically or chemically bound to the textiles. The determined TDS of the outgoing water from each washing cycle of the first wash are plotted in Figure 1. The measured TDS (0,4 mg/L) of the deionized wash water fed to the machine at the initiation of each cycle, and the reported upper limit (5 mg/L) of tolerable TDS,¹⁰ is included in the plot for comparison.



<u>Figure 1:</u> Observed levels of solutes in comparison to the levels of the feed water (0,4 mg/L) and a reported¹⁰ upper limit of the same (5 mg/L).

It is reasonable to assume, after introduction of feed water at the beginning of any cycle except the first cycle, that an initial rapid dilution of the solutes of the remaining water of the wet textiles from the previous cycle, will occur. During the washing cycle, the solute concentration will increase further due to solvation of previously undissolved materials of the textiles. Such materials may be classified as belonging to one of two groups, medium releasable materials, e.g. minerals and organic compounds with relatively slow dissolution kinetics, and low releasable materials, e.g. materials originating from the textiles *per se*. In the first washing cycle, when the dry textiles are initially exposed to water for the first time of the washing procedure, the corresponding easy releasable materials, e.g. group-I chlorides or urea from sweat, urine or the like, will be the major initial contributor of the solutes.

The observed TDS-values of the first wash agree with a substantially complete dissolution of all easy and medium releasable materials in the first two cycles, then followed simply by dilution of the

thereby resulting solute in the subsequent cycles. In the first cycle, the volume of washing water being substantially less than in subsequent cycles, a probable dissolution limiting saturation occurred. Thereby remaining undissolved materials explain the abnormally high solute concentration in the second cycle, in which these materials were free to dissolve as not limited by saturation.

An estimate of the contribution to the solutes of low releasable materials, i.e. the continuous leakage (C_L) from the textiles, can be derived from the last cycle in the second wash (Table 2, 8,7 mg/L), when correcting for the baseline contribution from the feed water (Table 1, entry 2, 0,4 mg/L). Hence, the thereby estimated C_L is calculated to be 8,3 mg/L (8,7 mg/L – 0,4 mg/L).

A model of the washing process, based on a constant dilution over cycles and a constant C_L contribution from the textiles as explained above, being predictive of the observed TDS-values to a high degree of satisfaction, is manifested through the following equation:

$C_{n+1} = (([C_{WW} * (D-1) + C_n]) / D) + C_L$

wherein

 C_n : Solute concentration (TDS, mg /L) of output water following cycle no. "n".

 C_{n+1} : Solute concentration (TDS, mg/L) of output water following cycle no. "n+1".

 C_{ww} : Solute concentration (TDS, mg /L) of the fed water.

 C_L : Continuous leakage (TDS, mg/L) from the textiles.

D: The dilution factor.

In Figure 2, the observed TDS-values are overlaid with the corresponding modelled values. The following settings: $C_{ww} = 0.4 \text{ mg/L}$, $C_L = 8.3 \text{ mg/L}$ and D = 3.32, and the first measured TDS-value (351 mg/L) as the first seeding C_n of the model, were used. The apparent dilution factor D was iterated to give a best fit versus observed values (minimized sum of squared differences). The observed value (247 mg/L) was treated as an outlier, as motivated herein above, and thus not included.



Figure 2: Observed and modelled levels of solutes, i.e. TDS-values.

Above model was then used to simulate scenarios in which the feed water has a 10 times higher concentration of solutes (TDS = 4 mg/L) as compared to the actual case of the study (TDS = 0,4 mg/L). Such a feed water may be produced by a relatively simple and cheap reverse osmosis process (see for example Table 1, entry 8-10), without the need of e.g. ion-exchange resins requiring disadvantageous periodic regeneration. As can be seen in Figure 3, a dilution factor D identical to the current study case (D = 3,32), results in a solute profile which is practically comparable to the current case, in which a feed water with a relatively high purity (TDS = 0,4 mg/L) was used. Setting higher dilution factors D (D = 4 and D = 5) would, according to the simulation, yield a profile which is even better than the study case. In practice, higher dilution factors D could be achieved by increasing the volume of the feed water or spinning the textiles longer in between cycles (to decrease residual water), or both.



<u>Figure 3:</u> Comparison between observed levels of solutes using a 0,4 mg/L quality feed water and the corresponding modelled levels, using a 4 mg/L quality feed water and various dilution factors (D).

In summary, this study revealed that the clothes and textiles washed in a typical operational washing machine of detergent free type, which efficiency is claimed to be due to interacting water of very high purity, not for any significant period of time were in proximity with water having a TDS below 16 mg/L during a washing program intended "for heavily soiled laundry". This should be compared to the generally accepted corresponding upper limit for "ultrapure water" of use in semiconductor applications (TDS = 0,0001 mg/L) and as taught in a relevant patent application (TDS = 5 mg/L).¹⁰ With exception for the last two washing cycles, from which the output water had a TDS of 21,7 and 16 mg/L respectively, the corresponding TDS-levels of the preceding three cycles were found to be within the range of solute levels of common tap- or drinking water (351, 247 and 42,7 mg/L). It is further concluded, on basis of a developed model, that similar solute profiles, as determined on the studied washing machine, may be achieved by use of a less pure feed water. Hence, the current system, comprising tandem reverse osmosis and ion-exchange units for the generation of water having a TDS of 0,4 mg/L, in practice and for this purpose may be replaced by simpler, cheaper and environmentally more friendly purification systems, e.g. a single reverse osmosis unit generating feed water of about 4 mg/L quality.

It may be concluded that the results of this study indicate that the laundry never was subjected to what may be considered as "ultrapure water", "very pure water" or even water which is of significantly higher purity than what may be found in some natural spring waters.¹¹ Such an incongruity, between the experimental results of this study and the current claim of extraordinary cleaning properties of "ultrapure" water or the like, is clearly warranting further and extended open research in this area. It is suggested and hypothesized by the author that any subjectively determined acceptable cleaning results, in this and other similar detergent free washing machines, may be explained by a combination of user bias and the employment of mechanically and rinse-wise optimized washing programs, rather than to any hitherto unknown inherent property of water.

The author considers conducting future studies directed toward the quantification of non-polar solutes in the present water samples. In accordance with the hypothesis under challenge, the level of such non-polar solutes is expected to show an inverse correlation with the total solute concentration of the output water from a detergent free laundry machine of relevant type.

Finally, on basis of the results of this study, of various personal observations and of the results of previous experimental study reports,⁹ the author proposes the following alternative hypothesis for consideration:

"There is no significant difference of practical relevance for laundry applications in inherent cleaning ability between the case of < 1 mg/L quality washing water and the case of 100-300 mg/L quality washing water."

Acknowledgements

The author wishes to thank Jenny Holmquist, Miljöstrateg at MKB Fastighets AB, for providing access to the laundry facilities being the subject of this study, Thomas Nilsson, for technical assistance, Wepo AB, for the provision of the conductivity meter used in this study and all private support directed to Openexperiment.se¹² for the enablement of open studies like the present one.

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