NOM increase in Northern European source waters: discussion of possible causes and impacts on coagulation/contact filtration processes

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Abstract A significant increase in the content of natural organic matter (NOM) has been observed in several surface water sources in Norway during the past 10–12 years. Similar observations are reported also from test sites in Europe and North America. Water works in the southern part of Norway have experienced a doubling and in some cases even a tripling of source water color levels during the last decade. Other important characteristics of NOM, e.g. the specific UV absorption (SUVA), have changed as well. The change in raw water quality increases the required NOM removal capacity in water treatment, and significantly affects treatment process selection, design and operation. However, the operational impacts on water treatment processes are poorly quantified. This paper addresses the observed NOM increase in parts of Europe and North America. Although the reasons are not fully understood or identified, possible causes are presented and discussed. Factors like climate change or variability, especially alteration in precipitation patterns and land-use, as well as reductions in anthropogenic sulfur loadings seem relevant. From pilot testing on relevant raw waters, major operational impacts of increasing NOM and SUVA levels on coagulation-contact filtration processes are identified and quantified. As an illustration, an increase in raw water color from 20 to 35 mg Pt L−1 increased the required coagulant dose, sludge production, number of backwashes per day and residual TOC by 64%, 64%, 87%, and 26%, respectively. In addition, hydraulic capacity and filter run time decreased by 10% and 47%, respectively.

Keywords Climate change; coagulation; filtration; NOM increase

Introduction
Natural organic matter (NOM) is a key parameter with respect to design and operation of water treatment processes. NOM may negatively affect water quality by color, taste and odor problems, and promote biological growth and corrosion in distribution systems. It may also pose a health hazard as increased NOM levels also imply increased levels of complexed heavy metals and adsorbed organic micro pollutants. Furthermore, there is a concern regarding the formation of potentially harmful DBPs (THMs, HAA, MX, etc.) during water treatment. The elevated NOM levels increase disinfectant and coagulant dose requirements, and have negative impacts on the stability and removal of particles and pathogens. In many cases, NOM will control the optimal coagulation conditions and coagulant requirements. The reported increase in NOM content in surface water test sites in Europe and North America (Skjelkvaale, 2003) may therefore have rather severe consequences for the drinking water industry. The changes in raw water quality may affect the drinking water supply systems in that more NOM removal plants have to be built, and modifications of water treatment process design and operation are required in existing plants. Since water treatment processes are able to remove only a fraction of the total NOM, elevated NOM levels may also lead to a deterioration of tap water quality. These quite severe implications are, however, not yet addressed or quantified to any great extent in view of the reported NOM increase.
The main objective of this paper includes a discussion of the reported increase in NOM levels in parts of Europe and North America, with its possible causes. Furthermore, the implications of increasing NOM levels on drinking water treatment are quantified experimentally. In specific, the major impacts on coagulation and filtration processes are quantified through pilot plant experiments on raw waters with three different NOM concentration and SUVA levels.

**NOM increase and possible causes**

The increasing NOM levels seem related to global warming and/or changes in precipitation patterns. Reduced acid rain may also be an important factor (Skjelkvaale, 2003; Vogt, 2003; Vogt et al., 2001, 2002, 2004). Color and UV absorption appear to increase more than TOC, implying an increase in SUVA levels as well. This is indicative of a change also in NOM characteristics and treatability. There is a strong covariance between intensity of precipitation and NOM concentration in the discharge from forested sites. The reason is that increased runoff intensities lead to increased discharge from the upper parts of the soil profile, which is rich in soil organic matter (SOM). Decreased retention time in lake waters may further cause decreased photochemical degradation of colored NOM, decreased microbial degradation, and decreased in-lake coagulation and sedimentation. Consequently these processes may lead to a covariance of TOC with precipitation and/or discharge (Figure 1) as well as a seasonal variation in NOM quantity and quality.

Increased temperature will furthermore accelerate the degradation of organic material in soil, thus possibly generating more NOM, at least in the short term. Increased winter temperatures in northern latitudes will change the balance between rainfalls and snow as well as soil frost duration and thereby the pattern of leaching and watershed runoff.

On this background, the shifts in color and NOM may mainly be explained by climate variability and/or climate change, including primary and secondary effects of global warming and changes in precipitation patterns (Evans and Monteith, 2001; Skjelkvaale et al., 2001a, 2001b). Although the covariance between precipitation and NOM in many cases

![Figure 1](https://iwaponline.com/ws/article-pdf/4/4/47/417458/47.pdf)

**Figure 1** Empirical correlation between TOC and discharge for different seasons at the Birkenes catchment in southernmost Norway
may dominate the picture, it does not solely explain all the observed changes in color and NOM. Also effects of land use changes, decreased acid rain, and other important factors seem relevant to the ongoing changes in NOM quality and quantity (Figure 2).

In the regions experiencing unprecedented NOM increases in Norway there has also been reported a significant increase in annual increment and volume of growing forest stock (Tomter, 2000) and an increase in the amount of mosses during the 1980 and 1990s (Økland et al., 2001). These changes may be due to differences in forest management, prolonged growing seasons with longer and milder falls, increased atmospheric CO₂ levels, and the fertilizing effect of airborne nitrogen deposition. In Norway there has also been an increase in the relative ratio of deciduous to coniferous forest during the last few decades. The NOM supply from deciduous litter, especially alder and birch, seems to be substantial during the first fall (Hongve, 1999). Thus there may be some potential for NOM control through improved watershed management, e.g. selective removal of deciduous forest species.

In the Czech Republic a 50% decrease of forest cover in the 1970 and 1980s was followed by a strong decrease in NOM concentrations in stream water. The NOM is now increasing along with reduced acid rain deposition as well as a reestablishment of forest cover and climate change (Grunewald et al., 2003; Grunewald and Scheithauer, 2003).

Many of the regions now experiencing increased NOM were in the past exposed to high levels of anthropogenic sulfur loading. During the 1970s and 1980s, a reduction in color in lakes was reported in the regions suffering from acid rain (Gjessing, 1970). This was mainly explained by the protonation of the humic acids, as a result of the deposition of strong mineral acids, rendering them less hydrophilic. Furthermore, increased ionic strength and elevated concentrations of sulfate, H⁺ and metal cations resulted in increased complexation, coagulation and precipitation of NOM.

Furthermore, nitrogen is accumulating in the terrestrial ecosystems due to the continued fertilization by deposition of airborne nitrate and ammonia, leading to a build-up of terrestrial biomass. A likely resulting increased nitrogen content and thereby a reduced C/N ratio of the soil humic matter will in the short term encourage decomposition and thereby production of NOM, leading also to more NOM in surface waters.

![Figure 2](https://iwaponline.com/ws/article-pdf/4/4/47/417458/47.pdf)

**Figure 2** Possible reasons for increasing NOM in surface waters. Modified from Forsberg (1992) and Liltved (2002)
The large spatial variation in the amount and properties of NOM is a reflection of the spatial variation in watershed land-use. High NOM concentrations are associated with drainage from peatland, shallow upland soils, and watersheds with a high land/water ratio, i.e. large soil pools of humus relative to mineral soil and short water retention times. Conversely, low NOM concentrations are found in watersheds with sparse vegetation, poorly developed organic soils and large lakes (i.e. small pools of soil carbon and long water retention times) (Löfgren, 2003; Kortelainen, 2003). Studies of NOM quality in five Nordic watersheds showed that spatial variation in NOM concentration was positively correlated with both molecular size and hydrophobicity (Vogt et al., 2004). Using multivariate statistics on a similar data set of NOM material from sites in southern Norway, Oug et al. (unpublished) found DOC concentrations to be the key parameter explaining most of the variation in NOM characteristics. Even variations in qualitative parameters like specific optical properties (e.g. SUVA) were to a large extent empirically explained by the variation in DOC concentrations.

Results obtained within the International Cooperative Programme on assessment and monitoring of acidification of rivers and lakes (ICP Waters) indicate a significant NOM increase in test sites and regions throughout Europe and North America (Skjelkvaale, 2003). Maps showing the locations of the ICP Waters sites and regions can be found in the same references. Regional trend results for DOC (as mg DOC L⁻¹ year⁻¹) along with the number of sites within each region are presented in Table 1. This long-term monitoring activity shows that 6 out of 10 ICP regions analyzed exhibit significant positive slopes for DOC.

Similar trends of increasing organic carbon have also been reported from the UK (Evans and Monteith, 2001), from the Nordic countries (Skjelkvaale et al., 2001b), elsewhere in Europe (Skjelkvaale, 2001a), and in the US (Stoddard et al., 2003). In Canada, the picture is less clear (Jeffries et al., 2003).

### Implications of NOM increase on the water industry

**Need for additional water treatment capacity**

Some water works in the southern part of Norway have experienced a doubling and in some cases even a tripling of the raw water color level during the past decade. The increase in NOM levels will, in addition to the apparent aesthetic problem, promote the formation of potentially hazardous DBPs and increase the levels of both inorganic and organic micro pollutants. More water works therefore have to include NOM removal unit processes in

### Table 1

<table>
<thead>
<tr>
<th>Region</th>
<th>Continent</th>
<th># sites</th>
<th>Dissolved organic carbon – DOC</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Median</td>
<td>Lower CI</td>
</tr>
<tr>
<td>Alps</td>
<td>Europe</td>
<td>0</td>
<td>–</td>
</tr>
<tr>
<td>East Central Europe</td>
<td>Europe</td>
<td>17</td>
<td>+0.06</td>
</tr>
<tr>
<td>Northern Nordic</td>
<td>Europe</td>
<td>7</td>
<td>+0.05</td>
</tr>
<tr>
<td>Southern Nordic</td>
<td>Europe</td>
<td>18</td>
<td>+0.08</td>
</tr>
<tr>
<td>UK/Ireland</td>
<td>Europe</td>
<td>6</td>
<td>+0.13</td>
</tr>
<tr>
<td>West Central Europe</td>
<td>Europe</td>
<td>12</td>
<td>+0.03</td>
</tr>
<tr>
<td>Maine/Atlantic Canada</td>
<td>North America</td>
<td>17</td>
<td>+0.04</td>
</tr>
<tr>
<td>Vermont/Quebec</td>
<td>North America</td>
<td>16</td>
<td>+0.06</td>
</tr>
<tr>
<td>Adirondacks</td>
<td>North America</td>
<td>48</td>
<td>+0.06</td>
</tr>
<tr>
<td>Appalachian Plateau</td>
<td>North America</td>
<td>9</td>
<td>+0.03</td>
</tr>
<tr>
<td>Upper Midwest</td>
<td>North America</td>
<td>23</td>
<td>+0.06</td>
</tr>
<tr>
<td>Virginia Blue Ridge</td>
<td>North America</td>
<td>3</td>
<td>–0.04</td>
</tr>
</tbody>
</table>
In order to comply with the current water quality regulations. Others will have to upgrade and/or optimize their existing water treatment processes. There may also be a shift in the relative competitive power and capabilities as well as the optimum design and operational characteristics of various water treatment technologies as a result of the change in raw water quality. Because most treatment technologies are able to remove only a fraction of the organic matter, increased organic matter residuals and increased biological growth in the distribution systems may also result from the increasing raw water NOM concentrations.

**Impacts on water treatment technologies**

The increase in color seems to be more pronounced than the increase in DOC, implying that other important characteristics of NOM have changed as well. Thus the changes in raw water quality may seriously affect water treatability by different technologies, treatment costs, water treatment process selection, as well as treatment process design and operation. Studying the quality of NOM using principal component analysis, Vogt et al. (2004) documented that the specific adsorption ratio \( \text{SAR}_{UV} = \text{OD}_{A254nm}/\text{OD}_{A400nm} \) is negatively related to molecular weight (MW). A stronger visible color (i.e. reduced SAR) is due to increased length of the conjugated double bonds and is as such also conceptually related to the MW. An increase in specific color in the visual light range (i.e. OD at 300–600 nm/mg C) as observed in the regions with increases in NOM therefore implies a larger MW of the NOM. A larger MW NOM is less difficult to remove by coagulation and filtration.

**Experimental testing of NOM treatability by coagulation and filtration**

**Contact filtration pilot experiments**

In order to identify and quantify the implications of increasing NOM on water treatability by coagulation and filtration, numerous pilot experiments were run on raw waters with different NOM concentrations and SUVA levels. The filters used were Ø144 mm dual media filters with 0.65 m of 0.8–1.6 mm anthracite or light weight clay aggregates (Filtralite\textsuperscript{®}) on top of 0.35 m 0.4–0.8 mm sand. Three raw waters, i.e. NOM levels were tested: RW15, RW30, and RW50 with numbers indicating color levels of 15, 30 and 50 mg Pt L\textsuperscript{-1}, respectively, thus covering the typical range of Norwegian surface waters.

The raw waters RW30 and RW50 were obtained from RW15 by adding additional NOM in the form of a highly concentrated regenerate solution from a local full-scale macroporous anion exchange water treatment plant. All raw waters were low-turbidity waters (<0.3 NTU). Typically, the DOC and SUVA levels for RW15, RW30 and RW50 were 2.4, 3.8 and 5 mg DOC L\textsuperscript{-1}, and 3.8, 4.3 and 4.8 L m\textsuperscript{-1} mg C\textsuperscript{-1}, respectively. The raw waters were further characterized (HPSEC) by relatively high molecular weight DOC (2–50 kDa), in line with results obtained also for other surface waters in Norway. More details on raw water quality, methodology and pilot plant used in the experiment can be found elsewhere (Eikebrokk, 1999, 2002a).

**Results and discussion**

In general, residual metal turned out to be a critical parameter with respect to coagulant dose requirements. A sub-optimum coagulant dosage yielded residual metal exceeding the Norwegian water quality standard/guideline value of 0.15 mg Al or Fe L\textsuperscript{-1}. Less coagulant was needed in order to comply with the regulations on residual turbidity (< 0.2 NTU), color (< 5 mg Pt L\textsuperscript{-1}), and TOC (< 3 mg L\textsuperscript{-1}).

Treating RW30, typical results obtained were: a minimum coagulant dose (as alum) of 1.8 mg Al L\textsuperscript{-1} (i.e. 0.4–0.5 mg Al mg\textsuperscript{-1} TOC), a sludge production of 8 mg SS L\textsuperscript{-1}, a filter run time of 11 hrs (at 7.5 m h\textsuperscript{-1}; no polymer; terminated by breakthrough), and a residual
TOC of 1.9 mg L\(^{-1}\). From the HPSEC results the molecular weight distribution of most Norwegian raw waters seems different from the more low molecular DOC normally found in many other countries, including Australia. Based on the given raw water quality characteristics, good treatability of Norwegian raw waters by coagulation should be expected. The results seem to confirm this expectation.

On the basis of numerous pilot coagulation–filtration experiments, the impacts on important process parameters of the increasing raw water NOM and color levels were modeled and quantified, i.e. how factors like coagulant dose requirement, sludge production rate, filter run time and treatment capacity, backwash water (BW) consumption, etc. were influenced by the raw water color level. A summary of some major findings is presented in Figure 3, where the values obtained with elevated raw water color levels are normalized to the values obtained with a reference color level of 20 mg Pt L\(^{-1}\). The example presented in Figure 3 is based on coagulation with alum without any use of polymer as filter aid. The rate of filtration was 7.5 m h\(^{-1}\), and the filter runs were terminated by breakthrough within the 2 m of head available in the pilot filter columns.

As illustrated by the examples presented in Figure 3, an increase in the raw water color level from 20 to 35 mg Pt L\(^{-1}\) will have rather severe operational consequences.

- An increase in the number of backwashes per unit time, and the amount of inadequately treated ripening water, by a factor of 1.87
- An increase in the coagulant dose needed, and in the amount of sludge produced by a factor of 1.64
- An increase in residual TOC in treated water by a factor of 1.26
- A decrease in water treatment capacity by a factor of 0.90
- A decrease in filter run time (breakthrough) by a factor of 0.53

From the results presented above, some major implications of the increasing NOM levels are rather obvious. Increased consumption rates of chemicals for NOM coagulation and pH adjustment, increased sludge production rates, decreased filter run times and reduced treatment capacity, factors that also illustrate well increasing costs of operation.

![Figure 3](https://iwaponline.com/ws/article-pdf/4/4/47/417458/47.pdf)

**Figure 3** Impacts of increasing raw water NOM content (i.e. color) on major coagulation and filtration process parameters (alum coagulation, no polymer, 7.5 m h\(^{-1}\) filtration rate) (After Eikebrokk, 2002b)
Furthermore, the increase in residual organic carbon can lead to increased microbial growth in distribution systems.

Filter ripening as well as breakthrough implies a deterioration of water quality. The decreased filter run times and thereby the increased number of filter runs per day may therefore negatively affect the hygienic safety level of the treatment process.

The results may also contribute to improving the basis for evaluations of optimum treatment process selection and modifications in process design and operation in regions experiencing increasing levels of NOM.

**Conclusions**

In the past decade a significant increase in NOM concentration has been identified in areas of Europe and North America. Over the years 1990–2002 some surface water works in southern Norway experienced a doubling or even tripling of their raw water color.

Although the mechanisms are not fully understood, it is clear that the observed increase in NOM content of several Norwegian surface waters is mainly linked to changes in climatic conditions. Among the major climatic factors are changes in precipitation and hydrological flow patterns, elevated temperature levels, milder winters and extended primary growth season. Changes in forest management practices are also contributing to increasing forest volume and increasing ratio of deciduous to coniferous forests. Other explanatory factors, e.g. changes in anthropogenic atmospheric S-deposition and land-use, are needed in order to account for the observed spatial pattern of NOM increases. The latter factors may be of greater concern as they are changing at a greater pace than the climate.

With respect to raw water treatability the greater increase in color and SUVA than in DOC is highly relevant. Spatial variation in NOM concentration seems positively correlated with molecular size and hydrophobicity. Although the NOM increase may require more NOM removal capacity, the above factors are indicative of improved treatability by coagulation. However, the occurrence and impacts of additional changes in NOM characteristics have not yet been addressed.

In order to quantify major operational impacts of increasing NOM, experimental studies were conducted in a pilot coagulation-contact filtration pilot plant on low turbidity (<0.5 NTU) surface waters with color and DOC concentration levels of 15–50 mg Pt L\(^{-1}\) and 2–6 mg L\(^{-1}\), respectively. The SUVA\(_s\) of the tested raw waters increased with color from 3.8 to 4.8 L m\(^{-1}\) mg C\(^{-1}\), in accordance with the increasing SUVA levels observed in raw water sources with increasing NOM levels.

The experiments demonstrated well some major operational consequences and implications of quite realistic increases in NOM level. As an illustrating example, raw water color from 20 to 35 mg Pt L\(^{-1}\) resulted in 87% increase in the number of filter backwashes per day and in volume of filter ripening water produced, 64% increase in coagulant demand and sludge production rates, 26% increase in TOC residuals, 10% decrease in water treatment capacity, and finally 47% decrease in filter run time.

In addition to the significant impacts of increasing NOM on water treatment process selection, design and operation, this may also imply more violations of current water quality regulations on NOM and DBPs, including increased organic carbon residuals and increased biological growth problems in distribution systems.

**References**


