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RESEARCH ARTICLE

Browning of Scottish surface water sources exposed to climate change

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Abstract

Levels of dissolved natural organic matter (DNOM) are increasing in our boreal watercourses. This is manifested by an apparent increase in its yellow to brown colour of the water, i.e., browning. Sound predictions of future changes in colour of our freshwaters is a prerequisite for predicting effects on aquatic fauna and a sustainable operation of drinking water facilities using surface waters as raw water sources. A model for the effect of climate on colour (mg Pt L^{-1}) has been developed for two surface raw water sources in Scotland, i.e., at Bracadale and Port Charlotte. Both sites are situated far out on the Scottish west coast, without major impact of acid rain, with limited amounts of frost, and with limited recent land-use changes. The model was fitted to 15 years long data-series on colour measurements, provided by Scottish Water, at the two sites. Meteorological data were provided by UK Met. The models perform well for both sites in simulating the variation in monthly measured colour, explaining 89 and 90% of the variation at Bracadale and Port Charlotte, respectively. These well fitted models were used to predict future changes in colour due to changes in temperature and precipitation based on median climate data from a high emission climate RCP8.5 scenario from the HadCM3 climate model (UKCP18). The model predicted an increase in monthly average colour during growing season at both sites from about 150 mg Pt L^{-1} to about 200 mg Pt L^{-1} in 2050–2079. Temperature is found to be the most important positively driver for colour development at both sites.

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Introduction

Many boreal surface water sources have had a distinct increase in colour throughout the past few decades [1–3]. In the past this increase was mainly due to the decline in anthropogenic acid rain deposition [4, 5]. At present the changes in climate (i.e., increase in growing season) and land-use (e.g., less outfield grazing), resulting in increased biomass (i.e., catchment Greening), are strong drivers for the ongoing Browning [6–9]. The link is that incomplete decomposition of the increased biomass leads to increased input of allochthonous natural dissolved organic matter (DNOM), often in complexation with increased levels of iron [10–13]. Both DNOM and ferric iron species compounds have strong abilities to absorb light in the blue

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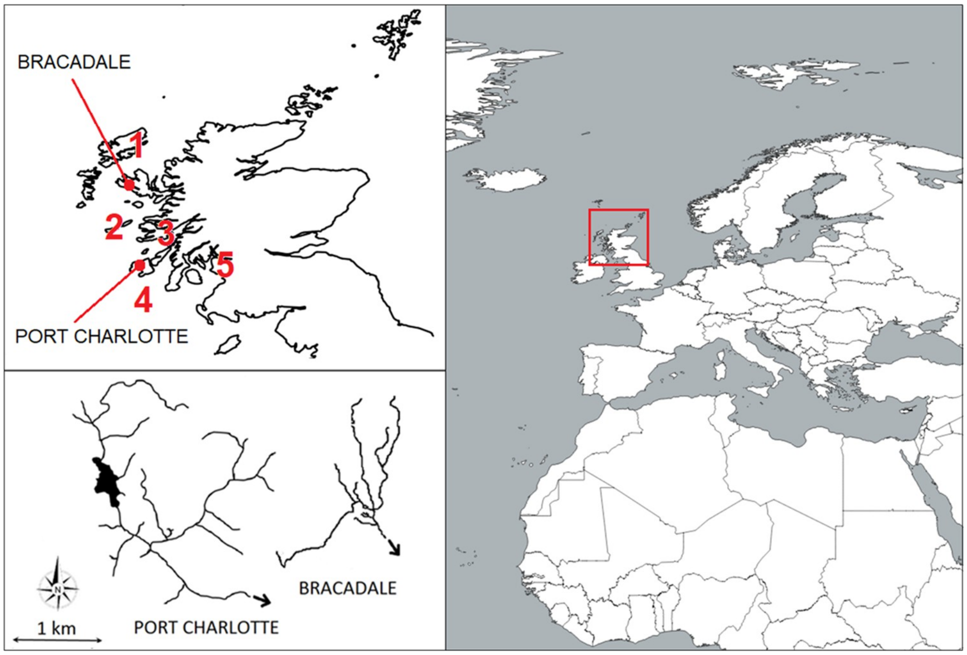

Fig 1. Raw water sources of Bracadale and Port Charlotte, located in the west of Scotland. Locations of meteorological stations (UK Met Office; historic station data) are shown with numbers 1–5 (mid figure); 1-Stornoway Airport, 2-Tiree, 3-Dunstaffnage, 4-Ballypatrick Forest (located in Northern Ireland), 5-Paisley. Maps are created in QGIS using shapefiles from Natural Earth (CC-BY 4.0; http://www.naturalearthdata.com). Stream networks for Bracadale and Port Charlotte (lower left; arrows indicate flow direction) are sketches made using satellite images.

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PAR-area [14–16], and Fe-DNOM complexes have been shown to enhance [17–19], but also to suppress [20] the DNOM absorbance. Increased colour and influx of DNOM has multiple effects on aquatic biota and represent a considerable challenge for drinking water treatment plants using surface waters as their raw water sources. An increase in DNOM concentrations may potentially require upgrading of waterwork facility, representing a substantial investment cost [21].

The large spatial and temporal variations in soils and the amount and quality of dissolved organic matter, governed by complex interaction with hydrological and microbial responses, is challenging to model. Here, a simple conceptually based but empirically fitted model-approach was chosen to link the main climatic driving factors to the fluctuations in colour.

This study presents a model for predicting near future changes in colour due to changes in climate at two small raw water sources; i.e., Bracadale and Port Charlotte. Both sites are situated far out on the west coast of Scotland. Bracadale is a pure lotic water system, whereas Port Charlotte also comprises a small water reservoir (Fig 1). Both sites are dominated by organic moorland [22].

Material and methods

Bracadale and Port Charlotte drinking water treatment plants are two small waterworks with highly coloured raw water sources, commonly reaching 150–200 mg Pt L^{-1} during the summer season (Fig 2). Variations in colour intensity are mainly due to annual fluctuations in concentrations of DNOM and to a lesser degree iron (Fig 2). Annual precipitation amounts are high with more than 2 000 mm yr^{-1} (Fig 3). Located on the western coast of Scotland, the catchments in this region have received lower levels of acid deposition in comparison to eastern

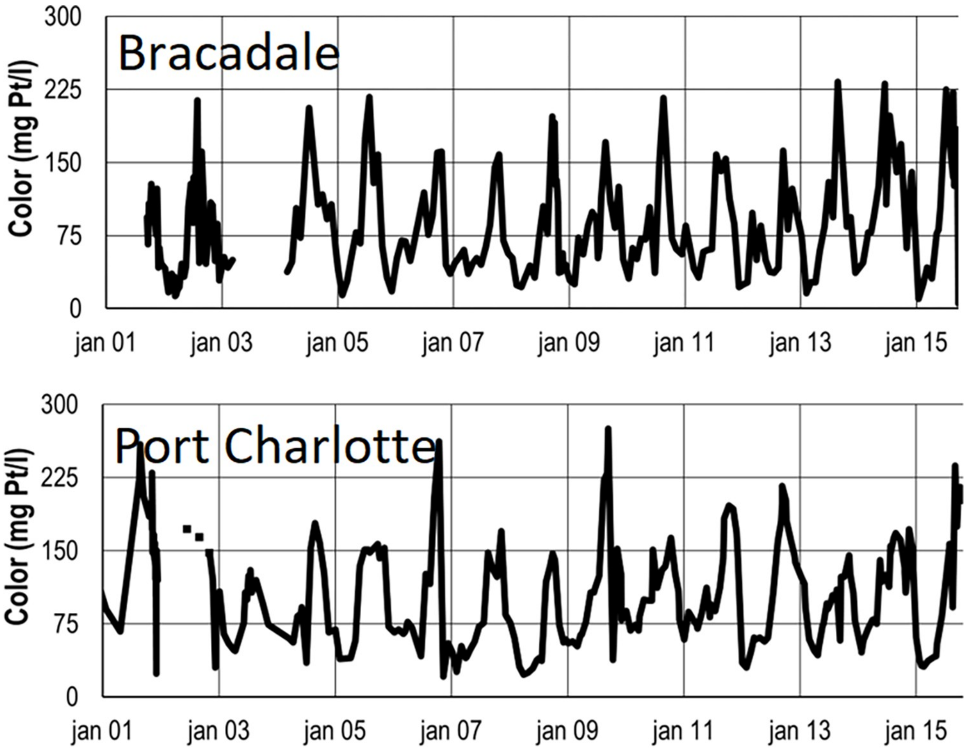

Fig 2. Long term data series on colour (mg Pt l^{-1}) (2001–2015), mainly on a weekly to bi-weekly temporal scale, sampled at Bracadale and Port Charlotte. Data provided by Scottish Water.

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regions of the UK [23, 24]. Minor long term annual changes in the electric conductivity (EC) of precipitation in the area are furthermore not expected to have led to any particular changes in soil DNOM concentration (low DOC_{prop}) [25]. Nevertheless, there has been a significant *percentage* decrease in the overall deposition of sulphur over the area from year 1986 to 2005 [24]. Conductivity of the raw water has been measured frequently at Port Charlotte from year 2000–2004 and is maintaining relative stable within the range of 100–150 μS/cm without observable indications of a downward trend. Furthermore, pH levels of the raw water have been measured at both sites throughout the entire duration of the studied period (year 2001–2016) and is mainly ranging between 6.5 and 7.5 at both sites with no observable indications of an upward trend. Due to the stable coastal climate air temperatures only rarely drop below 0˚C (Fig 3). From that, winter hydrology is more or less absent, and runoff would insignificantly be affected by freezing/thawing, snowmelt and ice-cover. The area has a significant history of land use, such as peatland ditching and sheep and goat farming. However, the land-use in the studied catchment sites would be restricted due to their designation as Drinking Water Protected Areas (DWPA) [26]. The Water Framework Directive ensures that no activities within DWPA areas lead to the degradation of water quality.

The model approach is based on a best fit parameterization using air temperature and precipitation amounts as conceptually based predictors for colour concentrations (Eq 1). A similar approach is presented and explained in Haaland *et al.* [6].

$$C_t = C_0 + k \cdot (Precip)^a \cdot [(T_{max} + T_{min})/2]^b$$

C_t is the modelled watercolour in mg Pt L^{-1}, while C_0 is the minimum background colour concentration (mg Pt L^{-1}) at each catchment. k is a constant adjusting for differences in

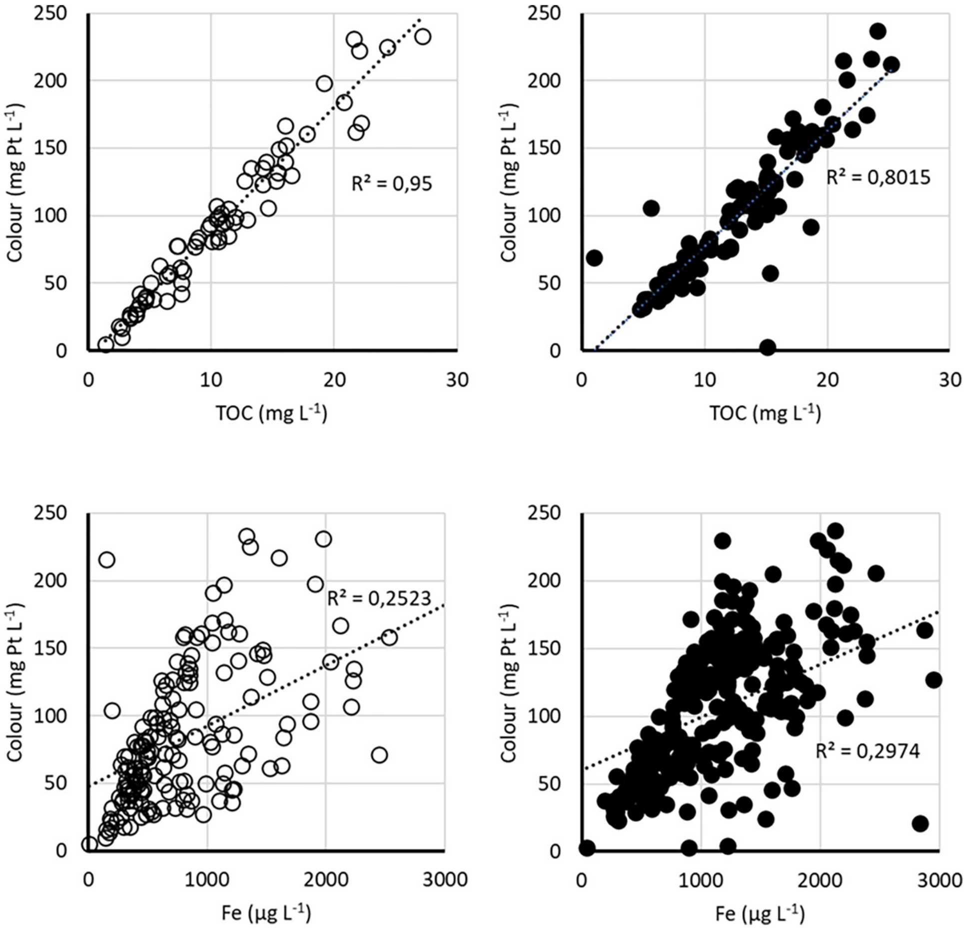

Fig 3. XY-plots for colour vs concentrations of iron (Fe) and total organic carbon (TOC, a proxy for DNOM) at Bracadale (open circles) and Port Charlotte (filled circles) for the period with accessible data; year 2000–2015. TOC (n = 68 at Bracadale and n = 106 at Port Charlotte) is here a stronger explanatory factor for colour than iron (n = 161 at Bracadale and n = 277 at Port Charlotte). Linear trend regressions: $r^2 = 0.80–0.95$ for colour vs TOC; $r^2 = 0.25–0.30$ for colour vs Fe.

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denomination (set to 1 for both sites). *Precip* is monthly amounts of precipitation (mm), which serves a proxy for runoff. *a* is a fitted catchment specific constant conceptually reflecting the impact of runoff on transport of DNOM from the soils to surface waters. Monthly weather data from five meteorological stations (Fig 1) were downloaded from the UK Met Office (Weather and climate change—Met Office). Meteorological data from the weather stations at Stornoway Airport, Tiree and Dunstaffnage were averaged and used for Bracadale, whereas averaged data from the stations at Tiree, Dunstafnage, Ballypatrick Forest (located in Northern Ireland) and Paisley were used for Port Charlotte. Precipitation amounts varied considerably between years, though there were no significant ($p < 0.05$) long term trends. As there are considerable spatial differences in the amount of precipitation between the weather stations the accuracy of this approach will at times be low. To adjust for absolute errors in precipitation amounts, the precipitation data were related to gridded (12 km^2) data for an area close to the sampling sites from UK Met Office database portal (https://climate-themetoffice.hub.arcgis.com/). These datasets are also for monthly average levels, though cover only the period 1991–2020. During this period the averaged weather data from the weather stations differed from the gridded data by a factor of 1,55 and 1,1 at Bracadale and Port Charlotte, respectively. These factors were therefore used to adjust the meteorological data. From this we expect that both precipitation amount and intensity have good accuracy and precision for both sites.

T_{max} and T_{min} are maximum and minimum monthly measured air temperature (˚C), respectively. Differences between the average temperature data from the Met UK meteorological stations and the Met Office database for T_{max} and T_{min} monthly averages at a nearby grid were small (often < 0.5 ˚C). The temperature data were hence not adjusted for deviation in levels. A few months in which minimum air temperature dropped sub-0˚C, i.e., 3 months at Port Charlotte and 1 month at Bracadale, out of a total of 180 months of data, were not used in the calibration procedure. Moreover, less days of frost are expected in the future according to the predictions from the UK Met Office [27]. The expression $(T_{max} + T_{min})/2$ was used. b is a fitted index conceptually adjusting for differences in the temperature effect on catchment production of DNOM. Increased temperature is expected to promote both increased primary production (catchment greening) and also higher DNOM concentration in surface waters due to increased microbial degradation of organic matter in the catchment soils. The temperature part of the model is thus a bulked proxy for temperature dependent enzymatic reactions [28] and can be regarded as a very simple exponential model fitted for an Eyring–Polanyi model for temperature dependence in biology [29].

The models were calibrated for a 15-year period of monthly average colour concentrations, based on weekly to bi-weekly data series, for each of the sites provided by Scottish Water through the NOMiNOR-project [22]. Data from odd years (2001, 2003, 2005, . . ., 2015) were used for calibration. A best-fit approach was used for optimizing the models in response to changes in climate (air temperature and precipitation amounts). Data from even years (2002, 2004, 2006, . . ., 2016) were used for a validation of the models. Differences in water balance (i.e., precipitation–runoff) due to differences in evapotranspiration on a monthly scale between years were by using this approach expected to be minor between the model calibration and validation periods.

Results

The model performance is shown in Figs 4–6. The constant describing the background (minimum) colour concentration, C_0, was fairly low at both sites but seldom less than 10 mg Pt L^{-1} (Fig 2). C_0 was thus set to 10 mg Pt L^{-1} at Bracadale and 20 mg Pt L^{-1} at Port Charlotte. Increased ionic strength has a strong impact on DNOM solubility [6, 25, 30] by decreasing the thickness of the diffuse double layer (DDL) [31, 32]. Decreased thickness of the DDL at higher ionic strength leads to increased DNOM aggregation and precipitation from solution. Although sea-salt (i.e., Na^+, Cl^-) is a dominating input constituent during stormy winter seasons, at both sites the runoff water during growing season is mainly constituted by calcium (Ca^{2+}) and bicarbonate (HCO_3^-). There are thus minor seasonal fluctuations in the ionic strength. The constant a is modulating the impact of precipitation. Based on the minor seasonal fluctuation a does not consider the effect of fluctuations in ionic strength on the solubility of DNOM. a has instead been interpreted as a best-fit constant reflecting the effect precipitation amounts has on the transportation of DNOM from the catchment soil solution to the surface waters. a was found to be 0.13 at Bracadale and 0.20 at Port Charlotte. The constant b, describing the non-linear effect of air temperature on colour concentrations, was best described at 1.6 and 1.5 at Bracadale and Port Charlotte, respectively. The level of change is somewhat comparable to what was found for phenolic compounds in laboratory experiments for UK peat soils [33]. A delay in climate response of one time-step (one month) was added to the Port Charlotte model because it contains a substantial reservoir pond inside its catchment (Fig 1).

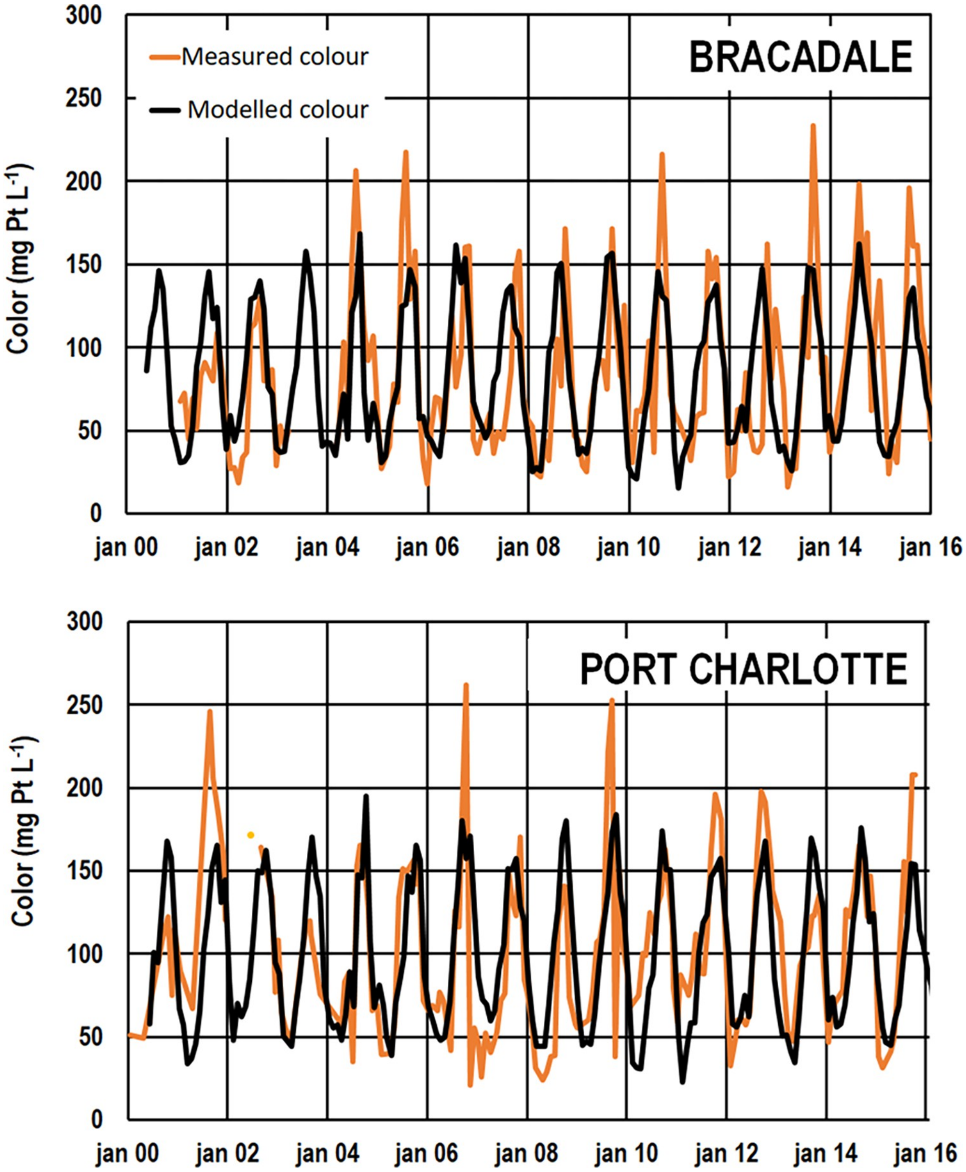

Fig 4. Model output (black lines) for colour at Bracadale and Port Charlotte, superimposed onto monthly averaged data (from existing 15 years of weekly to monthly measurements) of colour concentrations (orange lines) provided by Scottish Water. Modelled data is on a monthly scale. Measured data is on a biweekly to monthly scale. The highest uncertainty is for data sampled during late summer and early autumn, in where the most profound day to day variation in colour (mg Pt/l) are seen.

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Model performance

The model performance is good, though there are some deviations between measured and simulated data (Fig 4). Monthly colour values were well modelled (Fig 4), despite large variations on shorter time-steps, especially at the small lotic system at Bracadale, in comparison to Port Charlotte comprising a reservoir. Small headwater lotic stream systems have rapid direct response in water quality to changes in temperature and especially precipitation, whereas reservoirs integrate the terrestrial signal over a longer period. Moreover, the models were able to reproduce the seasonal fluctuations in measured colour concentrations with high accuracy on

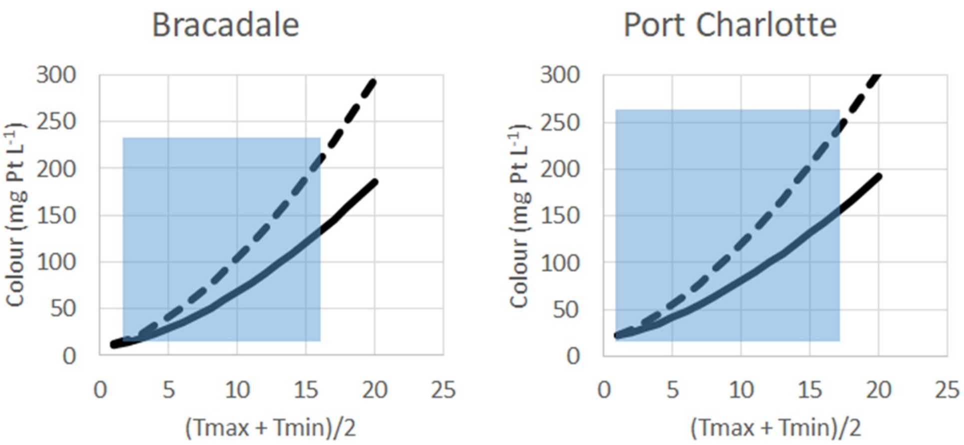

Fig 5. Modelled temperature response for monthly average colour development at Port Charlotte and Bracadale, shown for the minimum (solid black lines) and maximum (dotted black lines) estimated precipitation amounts (derived from nearby meteorological stations and UKCP Met portal). Blue squares depict the range of actual measured monthly average colour and temperature at both sites (2001–2016).

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monthly time steps (Fig 6). Linear regression between measured and modelled monthly averages of colour were strong; r^2 = 0.89 for Bracadale and r^2 = 0.90 for Port Charlotte. A likely cause for some lack of precision is the use of the mentioned spatially somewhat remote meteorological stations (Fig 1). Rain showers are particularly difficult to predict during growing season (at peak colour concentration), due to the large spatial and temporal variability in distribution of local scattered showers governed by convective airflows. Use of locally sampled meteorological data would therefore most probably have increased the model's precision and thereby its predictability.

Discussion

Effects of climate change, such as increase in air temperature and rainfall amount and intensity, are expected to have impacts on DNOM concentration and thereby colour of surface waters [6, 8]. Still, the effect of these changes in temperature and precipitation on export of DNOM from soils to surface waters will significantly differ between catchments. Sites dominated by thin soils or rock outcrops in catchments with high amounts of precipitation experience a high degree dilution of their runoff during rainfall episodes e.g. [34]. Such sites do typically not have an increase in DNOM concentrations in runoff during high discharge episodes and will thus not experience an increase in colour along with an increase in precipitation amounts due to climate change [6, 8]. However, throughout the model calibration period at the two studied sites, extreme episodes with high colour concentrations ($>$ 200–250 mg Pt L^{-1}) in the raw water were measured during late summer (Fig 2) characterized by heavy rainfall (Fig 7). This site-specific positive response in colour to increased runoff, implies that the runoff from these catchments is not being diluted (regarding DNOM) even during heavy rainfall events.

From the modelled temperature response for monthly average colour development (Fig 5) it can be deduced that temperature is an important positive driver for DNOM at both Bracadale and Port Charlotte. Likewise, several authors have found temperature to be a positive driver for concentrations of DNOM at some sites [30, 35]. Still, other sites are revealing it to be a negatively related driver i.e. [36]. Both lakes and streams are assessed in these studies. Lakes are more complicated and have biogeochemical processes that influence DNOM

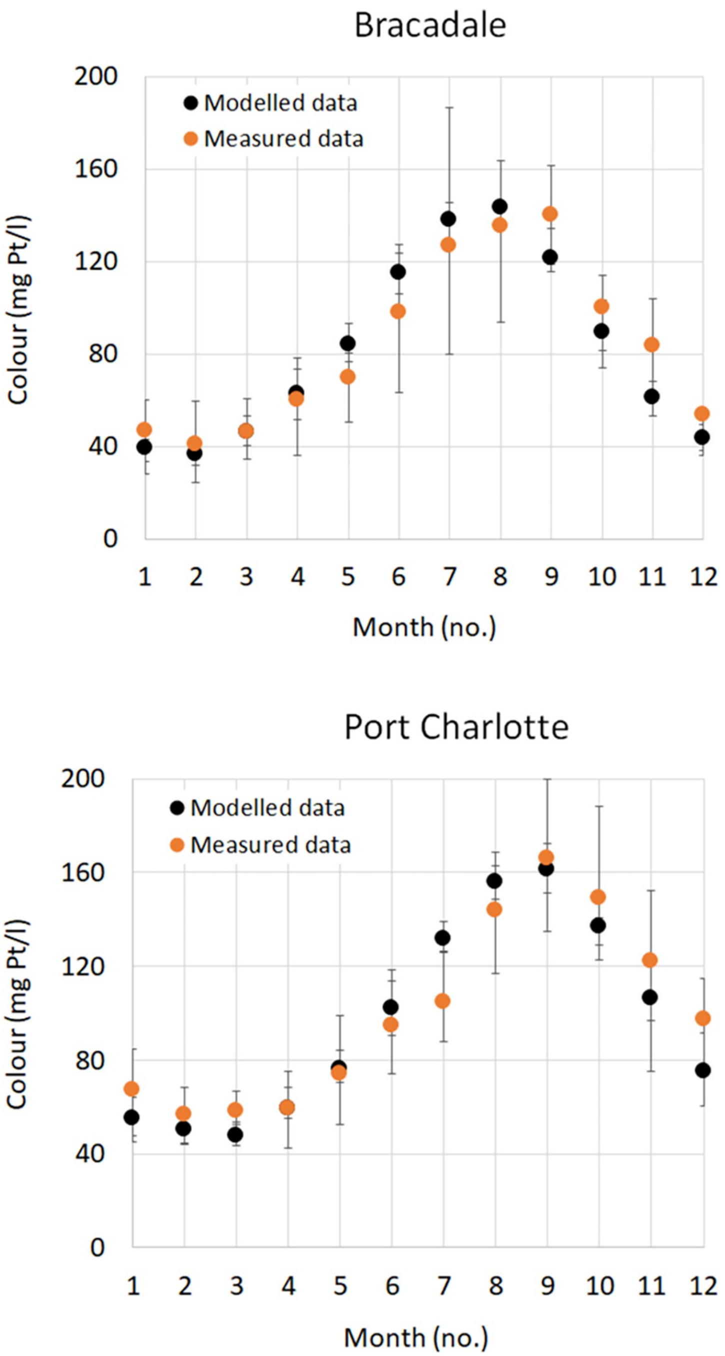

Fig 6. Monthly measured colour (orange circles) and modelled colour (black circles) averages, using monthly input data from year 2001–2016 at Port Charlotte and Bracadale (see text). Vertical bars indicate 25th and 75th percentiles. Linear trend line regression (r^2), between monthly measured and modelled averages are 0.89 for Bracadale and 0.90 for Port Charlotte.

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concentrations differently or more pronounced than streams. For instance, lakes' long retention times enhances the effect of microbial mineralization [37], as well as DNOM photobleaching and net sedimentation flux of DNOM. Shallow lakes often have several shorter vertical circulation mixing periods during growing season [38]. This might alter the distribution of DNOM concentrations in the water column from one day to another.

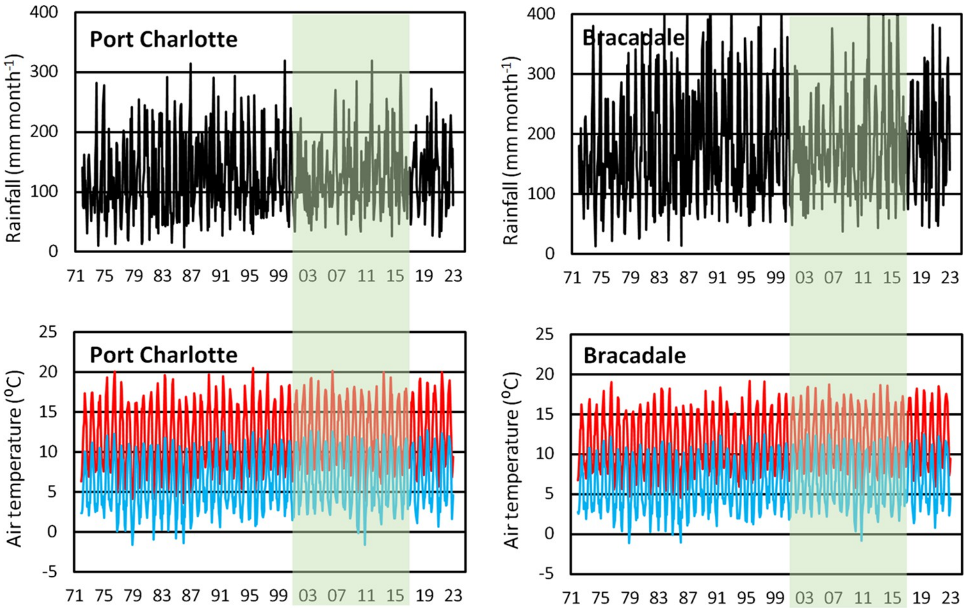

Fig 7. Long term data series on monthly average (1971–2023) on rainfall (*Precip*, mm/month) and air temperature (˚C, T_{max} (red lines) and T_{min} (blue lines) for Bracadale and Port Charlotte. Average measurements from daily sampled data for 2001–2016 at Stornoway Airport, Tiree and Dunstaffnage has been used as climate input data for Bracadale. Similar dataset from Tiree, Dunstafnage, Ballypatrick Forest and Paisley has been used as climate input data for Port Charlotte. Precipitation levels have been adjusted using gridded observation data from Met Office data base portal (https://climate-themetoffice.hub.arcgis.com/). The green squares indicate overlapping (mainly weekly to bi-weekly) chemistry data from Bracadale and Port Charlotte available for input to the model approach, provided by Scottish Water.

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The complicating hydrology of snow and ice (i.e., temperatures below 0˚C; Fig 3) and past acid rain deposition [23], as well as land-use changes, are minor issues at the Bracadale and Port Charlotte catchments. Furthermore, with a large and increasing surplus of runoff (> 2 000 mm rainfall yr^{-1}; Fig 3), the future catchments are not expected to dry up during growing season despite expected increased future temperatures. The amount of runoff from the catchments are instead expected to increase. Both temperature and rainfall are found to be positively drivers for concentrations of DNOM in the studied streams. The highest DNOM concentrations at Bracadale and Port Charlotte are during periods with higher temperatures during the growing season (Fig 2). High concentrations of coloured DNOM at Bracadale and Port Charlotte would hence most likely occur after warmer periods followed by an intense precipitation event in late in summer, transporting available DNOM from the catchment soils to the raw water surface recipients.

Climate change on future water colour development

Since air temperature and precipitation are the only dynamic parameters required in the colour models, they are well suited for predicting future changes in water colour governed by climate change. The scenarios produced for Eastern Scotland in 2018 [UKCP18) predict hotter summers as well as an increase in the frequency and intensity of extremes [27]. The UK Met Office uses the Hadley Centre Coupled Model version 3 (HadCM3) to make probabilistic downscaled climate projections for the UK. Future changes in air temperature and precipitation are based on predictions of several climate scenarios developed by the IPCC. A high emission climate RCP8.5 scenario for the period 2050–2079 with a 12 km^2 grid resolution, are

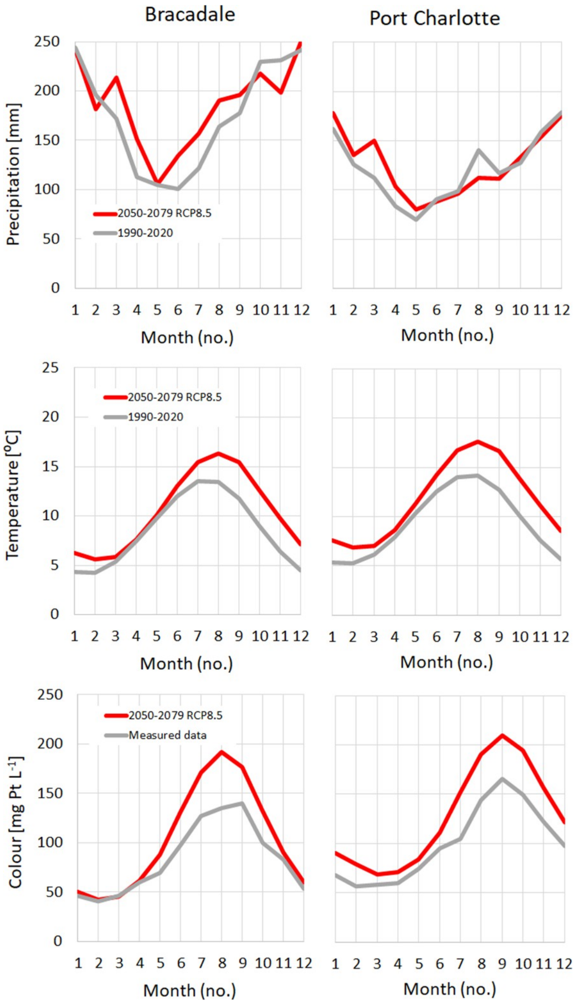

Fig 8. Climatic data from UK Met Office on monthly averages of air temperature and precipitation, using averaged gridded observed data (12 km^2) from 1990–2020, and also modelled data from the Hadley Centre Coupled Model version 3 (HadCM3) with median output using a RCP8.5 climate scenario. Grid number AD-49 from the Met Office climate data portal has been used for Bracadale catchment, whereas AD-34 has been used for Port Charlotte. Monthly averaged colour measurements (2001–2016) and modelled colour using the developed models for Bracadale and Port Charlotte, with inputs of predictions on temperature and precipitation from the HadCM3 model with the similar RCP8.5 scenario are also shown.

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available for the studied sites in the Met Office data base portal (https://climate-themetoffice.hub.arcgis.com/). Compared to the present climate data extracted from the UK Met data base portal and the meteorological stations, a median output form a run with the distinct RCP8.5 scenario, predicts an increase in temperature for the period 2050–2079 for Bracadale and Port Charlotte. The monthly average increase in temperature is between 2.5–3.5 ˚C during late growing season, autumn and winter at both sites (Fig 8). Distinct monthly average trends in precipitation amounts are not predicted to be significant at Bracadale and Port Charlotte (Fig 8). Based on these climate data our model predicts a monthly average increase in colour at both sites, especially during growing season. At Bracadale the model predicts an increase from less than 150 mg Pt L^{-1} to between 150–200 mg Pt L^{-1} as monthly averages from June to September. At Port Charlotte there is predicted a similar increase during the growing season, in addition to a general higher colour throughout the whole year (Fig 8). Increase in temperature is the most important driver at both sites. Extreme precipitation events are likely to become more frequent in Scotland in the future [27, 39]. Such episodes are not captured in these monthly average models. However, with increased storms in these areas situated near the ocean, it can be expected that there will also be higher concentrations of sea-salt (NaCl) in the precipitation. Higher concentration of sea-salt might decrease the solubility of DNOM in the catchment soils [25, 28]. However, at Bracadale and Port Charlotte changes in precipitation seems to be of less importance in governing DNOM concentration compared to temperature.

Conclusions

A model for effects of climate change on water colour has been fitted for two Scottish surface raw water sources. The model explains 89 and 90% of the variation in monthly averaged colour at the two sites. Using climate data scenario based on the RCP8.5 scenario, the model predicts an increase in monthly average colour during growing season from about 150 mg Pt L^{-1} to about 200 mg Pt L^{-1} in 2050–2079. Temperature is found to be the most important positively driver for colour development at the studied sites.

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Author Contributions

Conceptualization: Ståle Haaland.

Formal analysis: Ståle Haaland.

Methodology: Ståle Haaland.

Validation: Ståle Haaland.

Visualization: Ståle Haaland.

Writing – original draft: Ståle Haaland.

Writing – review & editing: Bjørnar Eikebrokk, Gunnhild Riise, Rolf D. Vogt.

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