Simulating Dissolved Organic Carbon Dynamics at the Swedish Integrated Monitoring Sites with the Integrated Catchments Model for Carbon, INCA-C

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Abstract Surface water concentrations of dissolved organic carbon ([DOC]) are changing throughout the northern hemisphere due to changes in climate, land use and acid deposition. However, the relative importance of these drivers is unclear. Here, we use the Integrated Catchments model for Carbon (INCA-C) to simulate long-term (1996–2008) streamwater [DOC] at the four Swedish integrated monitoring (IM) sites. These are unmanaged headwater catchments with old-growth forests and no major changes in land use. Daily, seasonal and long-term variations in streamwater [DOC] driven by runoff, seasonal temperature and atmospheric sulfate (SO_4^{2-}) deposition were observed at all sites. Using INCA-C, it was possible to reproduce observed patterns of variability in streamwater [DOC] at the four IM sites. Runoff was found to be the main short-term control on [DOC]. Seasonal patterns in [DOC] were controlled primarily by soil temperature. Measured SO_4^{2-} deposition explained some of the long-term [DOC] variability at all sites.

Keywords Dissolved organic carbon · INCA · Integrated Monitoring · Forest biogeochemistry

INTRODUCTION

Concentrations of dissolved organic carbon ([DOC]) in surface waters are changing in several parts of the world (Monteith et al. 2007), and long-term increases in [DOC] have been detected in many Fennoscandian locations (de Wit et al. 2007; Erlandsson et al. 2008; Lepistö et al. 2008). A number of hypotheses have been proposed to explain these trends, including climate change (Lepistö et al. 2008), declining acid deposition (Monteith et al. 2007; de Wit et al. 2007; Clark et al. 2010) and changes in flow and

rates of sulfate (SO_4^{2-}) deposition (Erlandsson et al. 2008). Moldan et al. (2011) have also recently demonstrated that rates of chloride (Cl^-) deposition significantly affect surface water [DOC].

Changes in [DOC] in surface waters may have important consequences for drinking water in Fennoscandia (Haaland et al. 2010). The costs of water treatment for human consumption and the production of potentially carcinogenic trihalomethane compounds are correlated with [DOC] (Christman et al. 1983). Changes in [DOC] also affect the timing and magnitude of thermal stratification in lakes, with consequent effects on ecological processes and mercury dynamics (Forsius et al. 2010). In addition, increasing [DOC] has been shown to affect pH recovery in many surface waters (Erlandsson et al. 2010).

Identifying causal relationships between potential environmental drivers and surface water [DOC] is confounded by the interacting effects of numerous factors. At many sites, while SO_4^{2-} deposition has been declining, the climate has warmed and runoff rates have changed. Geochemical modeling of the DOC dynamics in soil water at two of the Swedish integrated monitoring (IM) sites has identified a complex pattern of relationships between DOC and SO_4^{2-} deposition-driven changes in pH, ionic strength and aluminum chemistry, which can result in both increasing and decreasing trends in soil water [DOC] (Löfgren et al. 2010b).

A number of empirical and process-based modeling approaches have been used to explore the effects of potentially influential environmental factors on streamwater [DOC]. For example, stepwise multiple regression (SMR) has been used to predict interannual trends in [DOC] as a function of trends in SO_4^{2-} , Cl^- and temperature (Monteith et al. 2007). Short-term patterns in [DOC] have been simulated as a function of stream flow and soil

temperature (Köhler et al. 2009; Winterdahl et al. 2011 [this issue]). SMR of time-lagged climate and deposition parameters has shown that changes in acidic deposition can explain substantial proportions of long-term trends in surface water [DOC] in a series of Norwegian catchments (de Wit et al. 2007; Futter and de Wit 2008). End-member mixing analysis has been applied to simulate streamwater [DOC] as a function of inputs from catchment soils and wetlands (Easthouse et al. 1992), and a physically based model of DOC production and transport in boreal mires has been presented (Yurova et al. 2007). Detailed chemical modeling can also help to elucidate effects of soil water variables on surface water [DOC] (Löfgren et al. 2010b).

Regardless of the modeling approach used to simulate the behavior of DOC, understanding long-term changes in the environment requires high-quality long-term data. At the very least, long-term time series data on temperature, precipitation, atmospheric deposition of SO_4^{2-} , Cl^- and possibly nitrate, litter production, soil temperature and soil water chemistry, stream flow and streamwater chemistry are needed to understand the complex interacting effects of the numerous factors that influence surface water [DOC]. The Swedish IM sites are among the few locations where sufficient long-term data have been collected using harmonized protocols for sample collection and analysis, which facilitates comparison between sites and can aid in understanding environmental processes. IM networks and the data they generate in Sweden (Lundin et al. 2001), Finland (Bergström 1998), China (Tang et al. 2001), the UK (Morecroft et al. 2009), Canada (Vaughan et al. 2001) and elsewhere have been instrumental in understanding the effects of acid deposition on the environment (Forsius et al. 1998) and can play an important role in quantifying the effects of climate change.

In this modeling study, we evaluated the effects of climate- and deposition-related variables on streamwater [DOC] at the four Swedish IM sites using the INCA-C model. Specifically, we examined the effects of hydrology, modeled soil temperature and soil water [SO_4^{2-}] and measured SO_4^{2-} and Cl^- deposition on simulated streamwater [DOC].

MATERIALS AND METHODS

Site Characteristics

The four Swedish IM sites (Gårdsjön, Kindla, Aneboda and Gammtratten) are intensively monitored headwater and forested catchments located in protected areas (Lundin et al. 2001; Löfgren et al. 2011 [this issue]). Norway spruce (*Picea abies*) is the dominant tree species. Podzols developed in till are the dominant upland soil type at each site, while discharge areas are dominated by organic soils.

Large areas of exposed bedrock are present at both Gårdsjön and Kindla (Table 1). The Gårdsjön catchment is located near the west coast, has relatively thin soils and receives high marine inputs. There are three intensively monitored subcatchments at Gårdsjön. In this study, flow and DOC were simulated at the F1 control subcatchment. Aneboda is located in south-central Sweden and has deeper soils, while Kindla is located in a topographically steep area in central Sweden. Gammtratten is also located in a relatively steep area in northern Sweden. The stream at Aneboda only flows intermittently. All sites are recovering from acid deposition (Köhler et al. 2011 [this issue]; Löfgren et al. 2011 [this issue]).

Some of the most relevant site parameters for INCA-C modeling are presented in Table 1. Average air temperatures during the study period (1996–2008) were lowest at Gammtratten in the north and highest at Aneboda and Gårdsjön in the south. Annual average temperatures have increased at all sites (Fig. 1; Table 1). Precipitation was highest at Gårdsjön and lowest at Gammtratten. There has been a significant ($P < 0.05$) increase in precipitation at Kindla. Relatively little precipitation falls as snow at either Gårdsjön or Aneboda, but snowfall accounts for more than 20% of annual precipitation at Kindla and Gammtratten. Gårdsjön received the highest rates of atmospheric deposition of SO_4^{2-} and Cl^- (Fig. 1). Sulfate deposition has declined at all four sites (Table 1) but most rapidly at Gårdsjön (Fig. 1), while Cl^- deposition was highest and most variable at Gårdsjön (Fig. 1).

There were large differences in runoff between catchments (Fig. 1). Annual runoff ranged from an average of 340 mm year^{-1} at Aneboda to 570 mm year^{-1} at Gårdsjön during the study period. The seasonal distribution of runoff also differed markedly between the sites. At Gårdsjön, flows were highest in the winter months and low during summer, a typical pattern for sites in southern Sweden and other temperate environments. At Gammtratten and Kindla, there were low winter flows and spring snowmelt peaks, typical of boreal sites in Sweden and elsewhere. Seasonal runoff patterns at Aneboda were similar to those at Gårdsjön, but with a small spring snowmelt peak. The runoff patterns observed at Kindla and Aneboda depended on the winter snowpack, which determined whether the runoff regime was more similar to that of Gårdsjön (warm with no pronounced spring flood) or to that of Gammtratten (cold with a pronounced spring flood).

Streamwater sampled at Gårdsjön was filtered through a $0.45\text{-}\mu\text{m}$ filter before organic carbon concentrations were measured, while unfiltered water samples from the other three IM sites were analyzed. It has been shown that suspended matter does not significantly affect the organic carbon concentration in a large number of Swedish forest streams, including those at the four IM sites (Löfgren et al.

Table 1 Parameters at the four IM sites relevant for DOC modeling

Parameter	Aneboda	Gammtratten	Gårdsjön	Kindla
Latitude (N)	57°07'	63°51'	58°03'	59°45'
Longitude (E)	14°03'	18°08'	12°01'	14°54'
Elevation (m.a.s.l.)	210–240	410–545	114–140	312–415
Catchment area (ha)	18.9	39.6	3.6	20.4
Peat (%)	3	6	3	3
Thin peat (%)	9	4	0	0
Till, medium to high boulder frequency (%)	79	73	63	56
Isolated large boulders (%)	8	2	0	0
Thin till (%)	0	2	0	0
Bedrock outcrop (%)	<1	13	34	41
Land cover Type I: wet spruce and open mire (%)	18	14	11	24
Land cover Type II: dry forest (%)	82	86	84	71
Land cover Type III: other (%)	0	0	5	5
Annual air temperature, 1996–2008; average (min, max) (°C)	6.5 (4.3, 7.5)	2.4 (1.2, 3.3)	7.0 (5.7, 7.9)	5.2 (4.0, 6.0)
Temperature trend (Mann–Kendall <i>Z</i> & <i>P</i>)	2.69 (0.007)	2.68 (0.007)	1.95 (0.05)	2.68 (0.007)
Annual precipitation, 1996–2008; average (min, max) (mm year ⁻¹)	800 (590, 1000)	580 (420, 850)	1110 (830, 1330)	850 (700, 1130)
Precipitation trend (Mann–Kendall <i>Z</i> & <i>P</i>)	0.49 (0.62)	0.24 (0.81)	1.59 (0.11)	2.19 (0.03)
% of precipitation falling as snow, 1996–2008	8	29	7	21
Sulfate deposition trend (Mann–Kendall <i>Z</i> & <i>P</i>)	−3.78 (0.0001)	−3.13 (0.002)	−2.81 (0.005)	−2.68 (0.007)
Chloride deposition trend (Mann–Kendall <i>Z</i> & <i>P</i>)	0 (1.0)	−1.52 (0.13)	0.61 (0.54)	0.85 (0.39)
Annual average measured runoff (mm year ⁻¹)	340	480	570	500
DOC data availability	1996–2008	1999–2008	1996–2008	1996–2008
Average streamwater [DOC] (<i>n</i>) (mg l ⁻¹)	23.3 (275)	8.1 (233)	11.2 (255)	8.0 (291)
[DOC]:log ₁₀ (Runoff)	−5.23	2.02	1.10	−0.72
Volume-weighted [DOC] trend (Mann–Kendall <i>Z</i> & <i>P</i>)	1.10 (0.27)	−0.62 (0.53)	1.46 (0.14)	2.61 (0.009)

2010a), suggesting that unfiltered total organic carbon is a good proxy for DOC. Therefore, all measured concentrations are assumed to be DOC. Measurements of [DOC] in samples collected between 1996 and 2008 were available for all sites except Gammtratten, where observations began in 1999. Streamwater [DOC] were highest at Aneboda and lowest at Gammtratten and Kindla. Volume-weighted [DOC] significantly ($P < 0.05$) increased during this period only at Kindla (Table 1). Flow:[DOC] relationships can be used to characterize catchment behavior (see Winterdahl et al. 2011 [this issue]). Wetland-dominated sites typically display a negative flow:[DOC] relationship, while till-dominated boreal sites typically display a positive flow:[DOC] relationship. Flow:[DOC] ratios were positive at Gammtratten and Gårdsjön and negative at Aneboda and Kindla (Table 1).

Streamwater [DOC] can vary at daily, seasonal and long-term time scales. To quantify the relative magnitudes of daily, seasonal and long-term variations in [DOC] time series at the four IM sites, analysis of covariance (ANCOVA) was applied in which [DOC] at each site was modeled as a function of log₁₀(Runoff) and nominal variables representing the month and year in which an observation was made (Eq. 1):

$$[\text{DOC}] = \beta_0 + \beta_1 \log_{10}(Q) + \beta_{2,i} + \beta_{3,j} + \varepsilon \quad (1)$$

In Eq. 1, Q is daily runoff in mm day⁻¹, $\beta_{2,i}$ is the least squares estimate of [DOC] in month i and $\beta_{3,j}$ is the least squares estimate of [DOC] in year j (1996–2008). Relative contributions of short-term, seasonal and interannual components to the total variation in [DOC] at each site are shown in Fig. 2.

INCA-C Model

The Integrated Catchments model for Carbon (INCA-C) is a dynamic, process-based model for simulating DOC and inorganic (DIC) carbon dynamics in soils and surface waters. The model simulates the production, transformation and fluxes of carbon from soils to surface waters and operates on a daily time step. It has been applied to small, forested headwater catchments in Canada (Futter et al. 2007) and Fennoscandia (Futter and de Wit 2008; Futter et al. 2009) and to large, mixed land-use catchments in Canada (Oni et al. 2010).

In INCA-C, a catchment is simulated as a set of one or more land cover types, each represented as two soil boxes

Fig. 1 Hypothesized drivers of stream [DOC], including average monthly runoff for the whole period of observation (a), annual chloride deposition (b), annual sulfate deposition (c) and annual average air temperature (d)

with an overlying surface organic layer. Water percolating through the surface organic layer may incorporate organic carbon from the litter layer or humus before entering the upper soil box, where organic carbon sorption/desorption and mineralization to DIC occur. From the upper soil box, some water enters the stream and the remaining water percolates to a lower soil box where further organic carbon sorption/desorption and mineralization occur. All water entering the lower soil box eventually enters the stream. The model is described more completely in Futter et al. (2007).

Daily time series of air temperature, precipitation, soil moisture deficit (SMD) and hydrologically effective rainfall (HER) data are needed to run INCA-C. HER is assumed to represent the water that eventually contributes to stream flow and is calculated as precipitation minus interception, evaporation and transpiration. Daily estimates of the precipitation and snowmelt reaching the soil surface are obtained from HER. Observed air temperature and precipitation time series

from the IM meteorological sites were used to simulate both SMD and HER with the HBV rainfall runoff model (Bergström 1992; Sælthun 1996). In INCA-C, modeled runoff is controlled by HER inputs and estimated parameters representing water residence times in the soil boxes. Flow in INCA-C is simulated using daily time series of HER, calibrated soil and groundwater residence times and calibrated stream velocity parameters (Futter et al. 2007).

Using INCA-C, it is possible to simulate effects of hydrological, climate- and atmospheric deposition-related variables on organic carbon dynamics. All processes are simulated as a series of linked first-order differential equations. Key processes include organic carbon inputs through litterfall, sorption/desorption of organic carbon in organic and mineral soils, organic carbon mineralization and advective transport of material from soils to surface waters. All process rates are dependent on modeled soil temperature and moisture (Futter et al. 2007). Soil temperature is modeled from air temperature

Fig. 2 Proportion of variation in [DOC] at the four IM sites that could be explained by short-term ($\log_{10}(\text{Runoff})$), seasonal (month) and long-term (year) variability

using a relationship developed by Rankinen et al. (2004) in which soil temperature is controlled by air temperature, insulating effects of snow cover, a soil thermal conductivity coefficient, heat capacity due to freeze/thaw and modeled soil temperature on the preceding day.

Within INCA-C simulated process rates are faster in warmer, wetter soils. Process rate dependence on soil temperature is simulated in the following manner (Eq. 2):

$$k_T = k \times \eta^{T-\theta} \quad (2)$$

In Eq. 2, k_T is the process rate at soil temperature T ($^{\circ}\text{C}$), k is the base rate, η is a calibrated temperature-dependent rate multiplier and θ is a temperature offset. Both k_T and k have units of day^{-1} . Changes in the mass of organic carbon in a soil box are driven by changes in soil temperature (T), moisture (m) and the masses of DOC sorbed ($k_S\text{DOC}$, kg C day^{-1}) and soil organic carbon (SOC) desorbed ($k_D\text{SOC}$, kg C day^{-1}) (Eq. 3). The base rates of the desorption (k_D) and sorption (k_S) coefficients are estimated during model calibration.

$$\frac{d\text{DOC}}{dt} = m(k_D\text{SOC} - k_S\text{DOC}) \quad (3)$$

Three terrestrial pools of SOC are simulated in each terrestrial land cover class in INCA-C. Conceptually, these

pools represent “new” organic carbon produced from plant litter and “old” organic carbon in the organic (upper horizon) and mineral (lower horizon) soils. DOC can be produced from each of the SOC pools.

Effects of strong acid anions (SAA, SO_4^{2-} and Cl^-) on organic carbon sorption/desorption dynamics were simulated. Equations 4 and 5 are simplified representations of effects of SAA on organic carbon sorption/desorption rates. The full equations are described in Futter et al. (2007). Briefly, they have been modified to include the effect of soil water [SAA] on the rate at which DOC is precipitated to SOC. At higher [SAA], organic carbon in the soil will appear to be less soluble as the transformation of DOC to SOC becomes more rapid. Using a negative value of b_1 facilitates the simulation of competition between DOC and SAA such as SO_4^{2-} for exchange sites in the solid phase.

$$\frac{d\text{DOC}}{dt} = m(k_D\text{SOC} - (b_1[\text{SAA}] + k_S)\text{DOC}) \quad (4)$$

$$\frac{d\text{SOC}}{dt} = m((b_1[\text{SAA}] + k_S)\text{DOC} - k_D\text{SOC}) \quad (5)$$

Futter et al. (2009) used simulated SO_4^{2-} deposition as a surrogate for soil water concentrations to model the effect of $[\text{SO}_4^{2-}]$ on DOC solubility. Here, we ran three sets of calibrations using (i) simulated B-horizon annual average

soil water $[\text{SO}_4^{2-}]$ time series values (“Total A Sulfate”; Köhler et al. 2011 [this issue]) from MAGIC model applications to the IM sites (Köhler et al. 2011 [this issue]), (ii) measured SO_4^{2-} deposition (Löfgren et al. 2011 [this issue]) and (iii) measured Cl^- deposition. We hypothesized that using measured SO_4^{2-} deposition to represent soil water [SAA] would result in better INCA-C model fits than could be obtained with MAGIC-simulated $[\text{SO}_4^{2-}]$. As Moldan et al. (2011) have shown, an inverse relationship between Cl^- in soil water and streamwater [DOC], annual time series of Cl^- deposition were used to simulate the effects of Cl^- on streamwater [DOC] at all sites except Gammtratten, where the short time series of observed data precluded their use in model calibration. As deposition observations did not begin at Gammtratten until 1999, an empirical relationship between MAGIC soil solution concentrations and observed deposition between 1999 and 2008 was used to estimate SO_4^{2-} deposition from 1996 to 1998.

All HBV and INCA-C simulations were carried out for the period 1/1/1996–31/12/2008, and all catchments were simulated as a single reach. The temperature offset, θ (Eq. 2), was set to a value of 0 for all simulations. During modeling, all in-stream processing was turned off; that is, only soil processes were allowed to control the DOC dynamics, and no attempt was made to simulate the behavior of DIC. Soil temperature time series from 10 cm at Aneboda and 5 cm at the other three sites were used to calibrate INCA-C-modeled soil temperature.

Model calibration was performed in two stages. HBV was calibrated first. Once an acceptable HBV calibration had been obtained, it was used to generate time series of HER and SMD for running INCA-C. The fit of the INCA-C model to the data was assessed by statistical (see below) and visual comparison of modeled and observed soil temperature, stream flow and surface water [DOC]. Soil thermal conductivity parameters were adjusted until an acceptable visual match was obtained between modeled and observed soil temperature. Stream flow velocity, terrestrial water residence times and the base flow index were then adjusted to find the best possible fit between INCA-C-estimated and observed stream flows. Only then was an attempt made to fit the observed streamwater [DOC] time series. This was done by adjusting sorption/desorption rates, soil temperature and moisture-related parameters and terrestrial hydrological parameters. Calibrations were assessed using Nash–Sutcliffe (NS) and Pearson correlation (R^2) statistics comparing modeled and observed runoff and streamwater [DOC].

HBV was calibrated using observed air temperature, precipitation and stream flow time series. A preliminary manual calibration was performed to maximize the NS statistics comparing (i) observed to modeled and (ii) log-observed to log-modeled stream flows. This parameter set was then used as the basis for a Monte Carlo exploration of the parameter

space. A rectangular parameter distribution ranging between $\pm 25\%$ of the values obtained in the original manual calibration was sampled 10 000 times using a Latin hypercube sampling (LHS) scheme (Futter et al. 2007). Parameter sensitivity was assessed by retaining the best-performing 100 parameter sets and comparing the posterior parameter distribution to a rectangular distribution using a Kolmogorov–Smirnov statistic. Sensitive parameters had a non-rectangular posterior distribution. The best-performing parameter set from the Monte Carlo analysis was retained and subjected to further manual calibration. This parameter set was then used to generate SMD and HER inputs for INCA-C.

Previous experience with INCA-C has shown that splitting catchment land cover into two classes—one representing dry upland forests and the other wet forests, open mires or swamps—is conducive to reproducing observed dynamics in surface water [DOC] time series (Futter et al. 2007, 2009; Futter and de Wit 2008). In the simulations presented here, three land cover classes were used, which were based on land cover classes described in Löfgren (2009) (Table 1). Wet spruce forest and open mire comprised one land cover class. *Vaccinium myrtillus*-spruce forest, *V. myrtillus*, mixed conifer stands and young pine stands were combined to form a second land cover class, and all other land covers (identified as “other” in Löfgren 2009) were lumped into a third land cover class. INCA-C was parameterized using a combination of manual parameter fitting and automated parameter space exploration. In the manual calibration, we attempted to maximize NS and R^2 of the modeled and observed streamwater [DOC] time series while avoiding negative effects on the goodness-of-fit between modeled and observed streamflows. The automated parameter space exploration was conducted using an LHS sampling scheme. The best-performing parameter sets obtained during the LHS parameter space exploration were retained for further manual calibration. Parameter sets were discarded if they simulated a greater than 10% change in mass of SOC.

A simple “one-at-a-time” sensitivity analysis of the INCA-C parameter sets was also performed, in which parameter values from the parameter set with the highest NS at each site were systematically varied by 200 and 50% of their value. This relatively large perturbation was applied to clearly establish the relative sensitivity of the parameters, defined as $\text{NS}_{\text{Best}} - 0.5 \cdot (\text{NS}_{\text{Double}} + \text{NS}_{\text{Half}})$.

Trends in annual modeled and observed time series were assessed using the non-parametric Mann–Kendall (MK) test.

RESULTS

Variance partitioning from the ANCOVA modeling of streamwater [DOC] as a function of runoff, month and year

revealed some marked differences between the IM sites (Fig. 2). $\text{Log}_{10}(\text{Runoff})$ explained approximately 40% of the variation in streamwater [DOC] at Gammtratten but only about 5% of its variation at Kindla. Between 15% (Gammtratten) and 35% (Aneboda) of the variability in [DOC] could be explained by seasonal, month-on-month variation. Approximately 25% of the total variation at both Gårdsjön and Kindla, but less than 5% at Gammtratten, could be explained by long-term year-on-year effects. More than 40% of the variation at all sites remained unexplained using the simple ANCOVA approach.

The HBV simulations reproduced the observed runoff patterns at each of the four IM sites (Fig. 3). NS statistics for the correspondence between modeled and observed/log-transformed modeled and log-transformed observed streamwater [DOC] values were (0.56/0.61) for Aneboda, (0.67/0.65) for Gammtratten, (0.66/0.69) for Gårdsjön and (0.58/0.59) for Kindla.

The INCA-C soil temperature model reproduced the observed temperature dynamics at each site. In general, modeled and observed temperatures were within 0.5°C of

each other. However, it tended to predict overly cold soil temperatures for Gårdsjön during the winter (not shown). Average modeled monthly soil temperature was strongly correlated with monthly predicted [DOC] (β_2 from Eq. 1) with R^2 values of 0.92, 0.83, 0.97 and 0.8 for Aneboda, Gammtratten, Gårdsjön and Kindla, respectively. For each site, summer maximum soil temperature was over- and under-estimated in different years. While there was relatively little variation in soil temperature between sites, slightly warmer maxima were recorded at Gårdsjön compared to the other three sites. There was no consistent pattern in annual minimum temperature, although the number of years in which soil temperature fell below zero was highest at Aneboda and subzero soil temperatures were rarely observed at either Gårdsjön or Gammtratten. The coldest soil temperatures were recorded at Kindla during the winter of 1998/1999.

Using measured climate and modeled soil water [SO_4^{2-}] data, it was possible to obtain (R^2 /NS values) of 0.52/0.51, 0.44/0.43, 0.43/0.42 and 0.29/0.29 for Aneboda, Gammtratten, Kindla and Gårdsjön, respectively. Using measured

Fig. 3 HBV-simulated (blue lines) and observed (red dots) runoff (mm day^{-1}) for Aneboda (a), Gammtratten (b), Gårdsjön (c) and Kindla (d). Note the differences in scale on the plots: there were higher runoff peaks at Gårdsjön than at the other sites

SO_4^{2-} deposition data instead of modeled soil water [SO_4^{2-}] led to large improvements in the model fit at Gårdsjön but negligible improvements at the other sites. The (R^2 /NS values) modeled and observed streamwater [DOC] were 0.53/0.52, 0.44/0.43, 0.44/0.41 and 0.39/0.39 for Aneboda, Gammtratten, Kindla and Gårdsjön, respectively. Using measured Cl^- deposition instead of SO_4^{2-} did not lead to any further improvements in model fit.

The INCA-C model was able to reproduce the seasonal and long-term streamwater [DOC] dynamics at each of the IM sites (Fig. 4). While it did not reproduce the high summer [DOC] observed at all of the sites, it generally captured both the interannual variability and seasonal patterns in autumn, winter and spring.

There was more interannual variability in estimates of annual average streamwater [DOC] calculated from observed data than in the INCA-C-simulated values (Fig. 5). While both unweighted and volume-weighted observed concentrations showed similar long-term trends to the INCA-C simulated values, there were some differences in trend strength (Fig. 5). Aneboda was the only site where there was a statistically significant increase in

INCA-C-estimated streamwater [DOC] over the study period (MK $P = 0.001$).

The INCA-C-estimated DOC export fluxes were highest for Gårdsjön and lowest for Kindla (Fig. 6). There were statistically significant increases in INCA-C-estimated DOC fluxes over the study period for both Aneboda (MK $P = 0.002$) and Kindla (MK $P = 0.01$). The increase in INCA-C-estimated DOC flux at Gårdsjön was not statistically significant (MK $P = 0.07$).

The rate of organic carbon desorption from the solid phase in the upper soil box was the most sensitive model parameter for all catchment simulations (Table 2). Perturbing the soil water chemistry effect on organic matter sorption (b_1 in Eqs. 4, 5) had the greatest effect on model goodness-of-fit at Kindla. This parameter also affected the fit of the model to the Gårdsjön data but very little influence on goodness-of-fit at either Aneboda or Gammtratten. Varying the soil temperature response (η in Eq. 2) had a large effect on model performance for Aneboda, moderate effects for Gårdsjön and Kindla, but virtually no effect for Gammtratten. Model performance was not sensitive to rates of litter incorporation into dissolved or solid organic

Fig. 4 INCA-C-modeled (blue lines) and observed (red dots) streamwater [DOC] (mg l^{-1}) at Aneboda (a), Gammtratten (b), Gårdsjön (c) and Kindla (d). Note the different vertical scales

Fig. 5 Volume-weighted (*blue*), unweighted (*green*) and INCA-C-estimated (*dashed red*) annual average [DOC] at the four IM sites: Aneboda (a), Gammtratten (b), Gårdsjön (c) and Kindla (d). Note the different vertical axis scales

carbon and was only slightly sensitive to water residence times in the soil boxes.

DISCUSSION

The four IM sites are all unmanaged headwater forested catchments. There were large differences in [DOC] dynamics between them during the study period, which could be simulated as a function of hydrology, soil temperature and soil water chemistry. There were differences in the relative importance of short-term, seasonal and long-term variability in [DOC] at the four sites. The amount of variation that could be explained by flow was the highest for Gammtratten and lowest for Kindla. There were negative flow:[DOC] relationships at Aneboda and Kindla, suggesting that DOC is diluted at high flows. Aneboda has the lowest relative relief and has the most extensive peat coverage of the four catchments (Table 1), which may strongly influence streamwater [DOC] at this site. While the

percentage of peat and thin peat coverage at Gammtratten and Aneboda were similar (10 and 12%, respectively), the length of the stream that runs through deeper wetlands is shorter than at Aneboda. Thus, there could be a greater influence of the wetlands on streamwater [DOC] at Aneboda than at Gammtratten. This hypothesis is further supported by the negative flow:[DOC] relationship at Aneboda and the positive relationship at Gammtratten (Table 1).

The NS statistics for the HBV simulations were satisfactory but lower than a target value of 0.8, which can often be obtained for larger catchments. As the HBV-estimated HER is used to drive runoff in INCA-C, the lack of fit in HBV simulations might lead to poor fits of INCA-C simulated [DOC]. Aneboda, the site with the poorest HBV fit, has an intermittent stream, which is more difficult to model successfully than continuously flowing streams.

It was generally possible to simulate effects of hydrological, climate-related and deposition-related variables on streamwater [DOC] adequately at the four IM sites using INCA-C. Model simulations were poorest for Gårdsjön, the

Fig. 6 INCA-C-modeled annual organic carbon export fluxes for the four IM sites

Table 2 Results of sensitivity analysis in which outputs from the best-calibrated INCA-C model for each site were compared with outputs obtained with perturbed ($2\times$ and $0.5\times$) parameter values

Parameter	Aneboda	Gammtratten	Gårdsjön	Kindla
Soil water effect on DOC sorption	0.02	0.02	0.23	0.41
DOC production from soil	1.44	1.65	2.82	1.05
Rate of litter incorporation into SOC	0.03	0.00	0.00	0.01
DOC production from surface organic layer	0.03	0.00	0.07	0.01
Soil moisture threshold	0.17	0.00	0.25	0.01
Soil temperature response	1.46	0.01	0.36	0.16
Soil thermal conductivity	0.04	0.02	0.02	0.02
Upper soil box water residence time	0.04	0.06	0.05	0.04
Lower soil box water residence time	0.02	0.03	0.01	0.00

Results shown are the NS statistic from the best model run minus the average NS statistic from the perturbed runs

smallest catchment. The results indicated that streamwater [DOC] was most strongly affected by hydrological variables at Gammtratten, but hydrology was less important at Gårdsjön and Kindla. This is consistent with the findings by Winterdahl et al. (2011 [this issue]) that flow-related controls on [DOC] are stronger at Gammtratten.

There are at least three possible reasons for the poor INCA-C simulations of some summer [DOC] values: a

large fraction of the organic carbon may be in particulate form; small volumes of water with very high [DOC] may be leached; and the calibration strategy used here may be biased against fitting high [DOC] values. We did not detect any significant relationship between the light (420 nm) absorbance of corresponding filtered and unfiltered samples, indicating that no particulate organic matter was present in the unfiltered samples that could explain the

differences. There is anecdotal evidence from observations of boreal headwater streams in Canada and Norway that small volumes of highly concentrated DOC may be leached from riparian areas during summer, and the small-scale hydrological processes leading to this leaching are not included in the landscape-scale INCA-C conceptual model of the environment. In addition, the calibration strategy used here involves attempts to minimize the sum of squares between modeled and observed values. All observations are weighted equally, so the calibration is biased toward fitting the more frequent observations of low [DOC].

There were differences between the observed and INCA-C-simulated annual average [DOC] at the four IM sites. Part of this difference might be due to the fact that the INCA-C estimates are based on values for each day of the year as opposed to just the days where observations were made. Eimers et al. (2008) have suggested that volume-weighted concentrations should be used when estimating long-term trends. While unweighted average concentrations showed the highest interannual variability, they followed similar patterns to volume-weighted and INCA-C-estimated concentrations. The INCA-C-estimated concentrations are less variable than the measured concentrations, which is consistent with the finding that high summer [DOC] is not well simulated. The only systematic differences ($>5 \text{ mg l}^{-1}$) between monthly modeled and measured [DOC] values were seen at Gammtratten during the spring flood period (April–May) and Aneboda during summer months (July–September). Again, at Aneboda, the intermittent stream may be the cause of some of the large monthly variation observed during summer. In future applications of INCA-C for simulations of these two sites, separate calibrations for spring, summer and the rest of the year might reveal different mechanisms of DOC mobilization.

The sensitivity analysis (Table 2) showed that temperature-related controls on surface water [DOC] are at least as important as soil water chemistry-related DOC sorption in INCA-C. This is in accordance with the findings that both seasonal (temperature-related) and long-term (deposition-related) changes affect streamwater [DOC]. The model simulations were relatively insensitive to changing hydrological parameter values, but this should not be taken as evidence that hydrology is unimportant. INCA-C model results are driven by HBV-estimated HER, the timing of which is strongly influenced by the amount of precipitation falling as snow. In the boreal region, warmer winter temperatures will result in less snow and more rain. This will change the timing of HER, which may potentially have large effects on simulated DOC fluxes (Whitehead et al. 2006).

Sulfate deposition decreased and average air temperature increased during the study period at all sites. This makes it extremely difficult to separate the effects of individual drivers (Futter et al. 2009). Temporal patterns in

average air temperature were similar at all sites, the main difference being that northern sites were colder than those in the south (Fig. 1). While both SO_4^{2-} deposition rates and modeled soil water $[\text{SO}_4^{2-}]$ have declined at all sites, the declines only affected INCA-C-modeled streamwater [DOC] at Kindla and Gårdsjön.

Gårdsjön was the only site where use of measured SO_4^{2-} deposition led to noticeable improvements in INCA-C streamwater [DOC] simulations. This suggests that Gårdsjön is the only site where soil water SO_4^{2-} plays a major role in controlling streamwater [DOC]. Neither MAGIC-modeled soil water $[\text{SO}_4^{2-}]$ nor measured SO_4^{2-} deposition is necessarily equal to the true, unobserved, catchment average soil water [SAA], which may influence the production of streamwater DOC. However, it appears that measured deposition is a better estimator of the soil [SAA] in the INCA-C conceptual model, which influences SOC solubility.

INCA-C-estimated DOC export fluxes were highest at Gårdsjön and lowest at Kindla (Fig. 6). This is somewhat counterintuitive, given the large differences in average [DOC] among the four IM sites, but emphasizes the strong effect of seasonality in flow on element fluxes. Fluxes are higher at Gårdsjön, in part, due to more precipitation falling as rain, leading to greater runoff. There were statistically significant increases in INCA-C-modeled DOC export flux at Aneboda (MK $P < 0.001$) and Kindla (MK $P = 0.03$). The increase in the export flux at Kindla was matched by increases in both volume-weighted observed [DOC] and precipitation (Table 1). As the increase in INCA-C-estimated volume-weighted [DOC] at Kindla was not statistically significant, the increase in simulated flux must be due to increases in both modeled concentration and flow. There were no statistically significant monotonic, long-term trends in either volume-weighted or INCA-C-modeled [DOC] at Gammtratten or Gårdsjön.

The Rankinen et al. (2004) soil temperature model incorporated in INCA-C was adequate for all seasons except winter. Given the importance of winter soil temperature for soil and streamwater [DOC] in the following spring (Haei et al. 2010; Ågren et al. 2010), this is a potentially serious shortcoming. Thus, efforts should be made to develop a simple soil temperature model, similar to that used in INCA-C, but with greater skill for simulating winter soil temperature.

As with all models with a large number of parameters, INCA-C is over-determined. There are a potentially infinite number of parameter sets, each of which is capable of generating model outputs with the same statistical relationship to observed data. This so-called problem of equifinality (Beven 2006) has aroused intense debate among environmental modelers. Model equifinality has been interpreted as meaning there is more than one possible explanation for an observed phenomenon. Modeling

approaches in which the smallest possible number of parameters capable of describing a data set according to some arbitrary statistical criteria have much greater success in reproducing observed environmental time series than is commonly seen with INCA-C model applications (Futter and de Wit 2008). However, over-reliance on simple models and purely statistical criteria may lead to overly simplistic views of the environment. If nothing else, it can be argued that more than 30 years of research into surface water DOC, and its links to acidification, acidity, climate and hydrology (Krug and Frink 1983; Laudon et al. 1999; Erlandsson et al. 2008, 2010; Lepistö et al. 2008; Clark et al. 2010; Haaland et al. 2010; Haei et al. 2010; Löfgren et al. 2010b; Löfgren and Zetterberg 2011), has shown that numerous factors influence or covary with surface water DOC. So, we must conclude that, on the basis of our current understanding, reality is over-determined but that models can be a useful tool for environmental hypothesis testing and understanding.

CONCLUSION

Concentrations of DOC, stream flow and soil temperature dynamics at the four Swedish IM sites were simulated using INCA-C. The results indicate that the main factors affecting surface water [DOC] differ at the four locations. At Gammtratten, a site with pronounced seasonality due to snowmelt, [DOC] is primarily controlled by flow. Soil temperature-related controls on modeled DOC production were important at all sites. The long-term modeled trends in streamwater [DOC] and flux at Gårdsjön and Kindla were linked to increases in temperature and declines in SO_4^{2-} deposition. Further efforts are needed to relate modeled to observed soil water [DOC] and to improve the soil temperature model. Continued support of the IM sites is of the utmost importance if the environmental consequences of changes in atmospheric deposition and climate are to be understood and models improved.

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