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Abstract:	Local remediation measures, particularly those undertaken in historical mining areas, can often be ineffective or even deleterious because erosion and sedimentation processes operate at spatial scales beyond those typically used in point-source remediation. Based on realistic simulations of an hybrid landscape evolution model combined with stochastic rainfall generation, we demonstrate that similar remediation strategies may result in differing effects across three contrasting European catchments depending on their topographic and hydrologic regimes. Based on these results we propose a conceptual model of catchment-scale remediation effectiveness based on three basic catchment characteristics: the degree of contaminant source coupling, the ratio of contaminated to non-contaminated sediment delivery, and the frequency of sediment transport events.
Response to Reviewers:	We greatly appreciate the thorough second reading by the reviewer and his comments. Here is the list of responses: 1)Most of the changes recommended made were carried out and the text is 99% fine. The answer to R1 point 6 is identical to R1 point 7... though point 6 was more of a recommendation than a necessary change. (It would improve the paper though). This is interesting - so its apparent that the TIMING as well as the DISTRIBUTION is controlled by the geomorphology - or the shape of the catchment. I think this is really really interesting and should be flagged up more in the discussion. Its also a good answer to the issue I raised about using the stochastic data - you can make more of this timing finding I think. We truly apologise for the mistake in giving the same answer to two different questions. It was obviously an editing confusion. Now, we have added in Conclusions: “While floodplain geomorphology greatly

influences the distribution of contaminated hotspots in the catchment, it also contributes to the timing of contaminant release from the outlet. The striking differences between the Ystwyth and Naracauli rivers, both in rainfall frequency and intensity, and in geomorphological attributes produce high retention capacity of contaminants in the first and low in the second. This differential in retention translates into high delays in flush-outs in the Ystwyth and very rapid responses to contaminant release from the mines in Naracauli (Fig. 7). Thus, both spatial and temporal patterns of formation of hotspots are consequences of geomorphology, and provide differential potential recipes for remediation in terms of long versus short term management”

2)Page 6, line 3: What is a tactical model? I’m not sure I’ve heard of this term before!! Maybe you could use a different word or re-word the explanation slightly?

Changed to “complex”.

2)Page 11, line 6: Catchment of floodplain geomorphology?

Changed to “floodplain geomorphology”

We trust that the above corrections are acceptable

Modelling remediation scenarios in historically mined catchments

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Abstract

Local remediation measures, particularly those undertaken in historical mining areas, can often be ineffective or even deleterious because erosion and sedimentation processes operate at spatial scales beyond those typically used in point-source remediation. Based on realistic simulations of an hybrid landscape evolution model combined with stochastic rainfall generation, we demonstrate that similar remediation strategies may result in differing effects across three contrasting European catchments depending on their topographic and hydrologic regimes. Based on these results we propose a conceptual model of catchment-scale remediation effectiveness based on three basic catchment characteristics: the degree of contaminant source coupling, the ratio of contaminated to non-contaminated sediment delivery, and the frequency of sediment transport events.

Keywords: CAESAR landscape evolution model, TRACER, mine remediation, stochastic rainfall simulation, sediment-associated contaminant dispersal.

Introduction

Since the industrial revolution metal mining has produced large quantities of metal contaminated waste that exists as a legacy in many river catchments worldwide (see Lewin and Macklin 1987, Macklin et al. 2006, and references therein). The presence of large quantities of metal contaminated sediment and soil in river catchments can have chronic impacts on ecological and human health. The high fertility of floodplains means that they tend to be used intensively for agricultural food production, and this has important implications for food security and human health (Miller et al. 2004; Abrahams and Steigmajer 2003; Smith et al. 2009, 2010).

Metal mining waste commonly exists as tailings, either in a liquefied or a solid form, providing a point source of primary environmental contamination many years after production. Environmental contamination occurs as a result of acute (collapse of tailings ponds) or chronic (leaching, aerial dispersion or erosion of tailings or metal contaminated sediment) release of metal contaminated wastes. As a result, river channels and floodplains in many parts of the world are contaminated with metal-rich waste as a result of the pollution associated with mining activities (Macklin et al. 2006).

Acute releases of metal contaminated sediment, such as at the Aznalcóllar mine, in Spain in 1998 (Hudson-Edwards et al. 2003), the Baia Mare and Baia Borsa accidents in 2000, northwest Romania (Macklin et al, 2003), and more recently the collapse of the tailings pond at Ajka (Hungary), deliver large volumes of metal contaminated waste into the environment. The collapse of the Aznalcóllar tailings pond in Spain resulted in a release of 5 million m^3 of acidified metallic wasted and led to extensive ecological damage to the especially sensitive Doñana National Park (Meharg et al. 1999; Turner et al. 2008). Though the full scale of the Ajkai Timföldgyár alumina plant disaster affecting Kolontár and Devecser has not yet been realised, an estimated 700,000 m^3 of alkali metallic waste was discharged (Anton et al. 2012) causing the death of 10 people. Although clean-up operations can remove a large amount of the contamination after an acute release of metal contaminated wastes, inevitably a proportion of the released material will persist in the floodplains and channel sediments providing a secondary source of contamination.

In the absence of an acute contaminant release, floodplains in mining affected catchments can still be significantly polluted as a result of the chronic long-term release (often historical) of contaminated water and sediment from spoil tips, flooded mine adits and poorly managed ore processing operations. Sediment associated metals disperse downstream, particularly during flood events, and are then deposited on floodplain surfaces during overbank flows. Contaminated floodplains remain a diffuse source of contaminated material, with residence times of hundreds if not thousands of years (Lewin and Macklin 1987). During periods of high flows, river bank erosion can reintroduce large volumes of metal contaminated sediment back into river channels resulting in dispersal and redeposition downstream (Dennis et al., 2003). The contaminated floodplain hazard is often not recognised, and when it is its scale is often underestimated.

Therefore, an understanding of the long-term sediment deposition and remobilization of contaminated sediments and soils is required, on a catchment scale, in order to accurately assess and predict contamination levels and dynamics (Coulthard and Macklin 2003). Landscape evolution models (such as CAESAR) can be used to predict future soil erosion and landscape stability (Hancock et al. 2010). Macklin et al. (2006) identified that an understanding of the geomorphological and hydrological processes in metal contaminated river systems is key to their management, in particular, identifying key areas of concern and prioritising remediation strategies.

In most cases the elimination of contaminated material from river catchments is most commonly based on point-source control at a local scale, either as bioremediation or simply as physical remediation measures (Ellis 1995, Macklin et al. 2006, Carlon et al. 2008). In order to link local remediation measures with their regional consequences, the modelling of contaminant dispersal should focus particular attention on predicting the long-term dispersal (from point and diffuse sources) and deposition of contaminants at the catchment scale. Therefore, identification of the appropriate spatial and temporal scales of the processes governing contaminant sedimentation is a crucial task to undertake if remediation measures are to be effective over a wide range of space and time scales.

The aim of this study is to provide a generic model for catchment scale remediation that 1) focuses attention on the hydrological and geomorphological processes, 2) identifies those catchments with higher pollution risks, and 3) determines which remediation measures are most likely to significantly reduce pollution in channels and floodplains. To address this we focus on three case studies within European catchments of contrasting climates, but all subject to historical mining activities. A series of calibrated, numerical simulations are used to develop predictions for sediment-associated contaminant dispersal/storage both with and without remediation measures in place.

Material and Methods

Focus study sites

The three study catchments were selected because between them they encompass a range of river system types, have different durations/intensities of mining activity and all have potential for present day acute/chronic contaminant metal release. First, the 193 km^2 Ystwyth catchment drains into Cardigan Bay on the west coast of mid-Wales (Fig 1.). It has a long history of Pb/Zn metal mining dating back to Roman times, but peak mining activity occurred in the nineteenth century and had largely ceased by the early twentieth century. Second, the 576 km^2 Ampoi catchment in western Romania drains the Apuseni Mountains and is a tributary of the Mures that drains into the Tisa/Danube drainage basin (Fig. 1). In contrast to the Ystwyth, mining activity (Cu, Zn, Pb) ceased less than 10 years ago and there are a number of major point sources of metal contamination in the form of three large tailings ponds and a smelter located at Zlatna (location in Fig. 10) in the upper part of the catchment. Third, the 30.2 km^2 Naracauli catchment drains the west coast of Sardinia. It is markedly different to both the Ystwyth and Ampoi catchments in that the catchment is relatively steep with a poorly developed floodplain, river flow is seasonal and mining ceased in the mid 1960s. However, there are remnants of several large unremediated tailings ponds and mine spoil tips that are actively eroding; contaminated sediments are transported downstream where they are deposited either on the Piscinas beach or flushed into the Mediterranean.

Simulating contaminant dispersal

CAESAR, a landscape evolution cellular automaton model, was used to simulate and predict the dynamics and distribution of metal contamination in catchments affected by past and present active mining activities. CAESAR calculates fine-scale sediment erosion and deposition processes over long, centennial-length time scales (Coulthard et al. 2000, Coulthard 2001), and coupled with TRACER it can model sediment-associated contaminant dispersal and deposition (Coulthard and Macklin 2003).

CAESAR and TRACER require the following quantitative information about the main external drivers controlling the movement and deposition of sediments and contaminants: hourly rainfall, hillslope and channel grain size distribution, a high resolution digital elevation model, the location of contaminant sources, and mine ore production data (Fig. 2). During the simulations, only grain size fractions corresponding to silt (<63 µm) and sand (0.063-2mm) are assumed to be susceptible to erosion and deposition. Validation of the model simulations was accomplished through correlation tests between distance profiles comparing simulation results with field data.

The model was run for the three catchments on a topography derived from an ASTER GDEM1 digital elevation model (DEM). CAESAR model runs were initiated either at the onset of large-scale mining or when reliable mining yield data were first available. Historical data were collected from the Ystwyth and Naracauli rivers based on the existing literature (Bick 1978, 1996; Progemisa 2002) thus permitting model validation with field data. In these catchments, mine locations were identified from ground survey and from Google Earth. The grain size distributions of hillslope and channel sediments were taken from field measurements. In this study we focus our results on the outputs for the most geochemically significant finest particle fraction (<63 µm). A lack of historical mining data for the Ampoi did not permit validation of past simulations with field data. Therefore, initial conditions for simulation runs in 2010 (and up to 2100) in the Ampoi were based on spatial kriging of sediment field data in the river, floodplains, and tailings.

Field data

In order to understand the distribution of the metal contaminated sediments on a catchment-scale and to validate the model outputs, detailed field surveys were completed in each of the three catchments. River and floodplain sediments were sampled every 900m on the Ystwyth and Ampoi rivers and every 90 m on the Naracauli; this shorter sampling interval was adopted because of the small catchment size and it provides an opportunity to validate the model over very fine spatial resolutions. Each sediment sample was air dried and sieved out at two size fractions (< 2 mm and < 63 µm) to reduce the effects of moisture content and sediment size variations on the subsequent XRF analyses. Niton Xlt hand-held XRF measurements of Pb concentrations were determined on both size fractions. XRF readings were recorded for a minimum of 1 minute; if the analytical precision was below 10% then readings were stopped at this point, if not, then readings were taken until the precision dropped below the 10% threshold.

A stochastic rainfall generation module

CAESAR requires hourly rainfall data, however, historically reliable and high-resolution temporal data for rainfall were absent or limited in all three catchments. Hourly data were only available from very few stations, often distant from the point of interest, at different elevations or in topographically contrasting areas, and frequently of short duration (typically 4-5 years). Furthermore, the production of hourly data are far beyond the capacities of current climate change models. Hence, a new module of stochastic rainfall generation has been developed to simulate hourly rainfall over centennial timescales. Based on the Modified Bartlett-Lewis Random Pulse model (MBLRP) (Rodriguez-Iturbe et al. 1988, Verhoest et al. 2010), a set of optimization procedures were used generate such a series, which in turn feed the rainfall input necessary to reliably run CAESAR/TRACER.

The modified Bartlett-Lewis model is a stochastic point process rainfall model. It creates synthetic data series when accurate and high-resolution rainfall events are needed. It modifies the original Bartlett-Lewis Random Pulse model (BLRP) by adding one parameter to the original list of five to better reproduce the probability of days with zero rainfall (Rodriguez-Iturbe et al. 1988). In the clustered model, rainfall is produced by cells of certain intensity that group themselves in storms (Koutsoyiannis and Onof 2001). Storm and cell arrivals t_i and t_{ij} follow Poisson distributions at rates

λ and β. Cell generation ending time within a storm v_i, cell duration w_{ij} and cell intensity follow exponential distributions with means γ, $1/\eta$ and μ_{ij}, respectively. η, in turn, is a parameter with a gamma distribution with a shape parameter α and scale parameter θ. Hence, the model presents six parameters in total (λ, β, γ, μ_{ij}, α, θ).

Theoretical expectations for the different statistical moments in the model were established by Rodriguez-Iturbe et al. (1988). The model, which is able to predict scale-dependent moments of the rainfall distribution, defines expected values for mean, variance, auto covariance, and days with zero rain probability. Parameters are found by minimizing a cost function by the method of moments (Verhoest et al. 1997), which minimizes the residuals between the monthly moments (required for stationarity of data, especially in highly seasonal climates) derived from a time series, and the theoretical expectations. The procedure requires the use of nonlinear constrained optimization procedures (Vanhaute et al. 2011) in a four-step procedure:

1. *Rainfall data series acquisition*: In order to accurately calculate the monthly moments of rainfall distribution, a long data series is required. Further, rainfall may vary widely spatially, so data series corresponding to the specific area to simulate are also needed. Simulated data series from the CIRCE (Climate Change and Impact Research: the Mediterranean Environment) European project were obtained. The set of simulations corresponded to the ERA40_1 scenario (control simulation), and the spatial resolution is 0.1 decimal degrees.

2. *Calculation of moments*: Due to the scale-dependent nature of rainfall distribution, moments were calculated at four different temporal resolutions (1 day, 2 days, 4 days, and 8 days). The seven moments selected corresponded to the mean, variance, covariance, autocorrelation functions at lags 1, 2, and 3, and the proportion of dry days. This provides a monthly matrix with 7 moments x 4 time resolutions to be used for optimization and downscaling to hourly resolutions.

3. *Determine MBLRP parameters by optimization*: The process of minimization of the cost function has been developed by implementation of a non-linear constrained optimization function. Due to the presence of multiple minima in the cost function, optimization was carried via the use of global optimization algorithms. A relevant constraint allowed a more accurate representation of extreme rainfall events: we set the constraint $\eta > 0.9$ (Verhoest et al. 2010). Imposition of the constraint and a good relationship between accuracy and computational speed triggered the choice of the pattern search algorithm (Hooke and Jeeves 1961) (Global Optimization Toolbox for Matlab 2009b) with a complete poll search method as the most ideal optimization procedure for this case. This algorithm is particularly effective at finding global minima in non-smooth parameter landscapes.

4. *Stochastic rainfall generation*: Once the optimum set of parameters is found, the simulated data series to be used in the CAESAR simulation runs is finally produced via a stochastic generation of the MBLRP model. The model produces strikingly realistic patterns of rainfall for all sites, from the distribution of the driest months (May and June for the Ystwyth, July and August for Naracauli, September and October for the Ampoi, Fig. 3) to the regularity of precipitation. The cumulative number of rain cells follows a linear response in the Ystwyth, indicating regular rainfall events across the year (Fig. 3a, upper panel). In Naracauli, however, rainfall cell events increase substantially in August, while the number of storms has two major increasing events, April, and September, matching the spring and autumn precipitation patterns (Fig. 3b, upper panel). Intense rainfall events are also present in Naracauli, with maximum hourly intensities larger than those in any of the other catchments. In the Ampoi river, both the storms and rains increase in June and November (Fig. 3c, upper

panel), and show irregular patterns of rainfall distribution across the year.

As a stochastic model, predictions arise from probabilistic outputs. That is to say, ideally the model should be run many times to produce confidence intervals for the predictions (Coulthard et al. 2012). However CAESAR is a complex model already incorporating no less than 25 parameters which should ideally be drawn themselves from bounded realistic distributions. This makes the task unrealistic for the aims of this study: to provide a conceptual model synthesizing the main geomorphologic drivers of remediation. Furthermore, over the large time scales involved in this study (300 years), the predictions of hotspot locations do not differ qualitatively at the end of such period. Hence we only incorporate a comparative picture with the results from single simulation runs for each catchment.

Results

Field data

Figure 4 shows downstream variations in stream sediment Pb concentrations in the three study rivers. On the Ystywth, Pb values show an initial exponential decrease from the Cwmystwyth Mine (c. 10 km) but then values remain relatively stable (1000 mg kg^{-1} +/- 500 mg kg^{-1}) for the remainder of the 35 km study reach. On the Naracauli there is no systematic downstream change in Pb levels but the values remain consistently high (typically > 5,000 mg kg-1) throughout the 8 km study reach. These metal levels are significant because 90% of the samples on the Ystwyth, and 100% of the samples on the Naracauli, exceed both the Dutch intervention (530 mg kg^{-1}) and the more stringent UK CLEA (450 mg kg^{-1}) values for Pb, indicating that these sediments are severely contaminated. Pb concentrations on the Ampoi are significantly lower than on the other two study river (Fig. 4) by one to two orders of magnitude. However, concentrations are elevated around the mining town of Zlatna (c. 10 km) and at the city of Alba Iulia (c. 40km), confirming previous observations that metal levels in river channel sediments in urban areas can often exceed those found downstream of mining facilities (Bird et al. 2008).

Model validation

Model validation was achieved by forward simulations of CAESAR/TRACER that incorporate the historical production of mine waste up to the field sampling year (2010), and comparing both the results in the field and simulations in terms of Pb concentrations along the main stream and river networks of the study catchments. The main output of TRACER is not a probability distribution per point in space and time, but rather the thickness of the contaminated sediment layer along the river and tributaries and the valley bottom. This can be transformed into actual soil concentrations because contaminants mix with soil particles in the different soil layers. Because of the lack of available historical mining data in the Ampoi, we have just performed CAESAR/TRACER validations in the Ystwyth and Naracauli.

The results for 2010 show very good agreements in the Ystwyth catchment between the simulated concentrations and the reported concentrations in the field (Fig. 5a). Concentrations peak around 33000 mg/kg just immediately downstream from the Cwmystwyth mining complex source and show a steep decline immediately afterwards, with high concentrations of around 5000 mg/kg in the floodplain around Cwmystwyth village, and another specific peak around 3000 mg/kg at the beginning of the floodplain downstream from the Grogwinion gorge, indicating those points as potential hotspots. In contrast, simulations in the Naracauli generally underestimated observed Pb concentrations along most parts of the river channel and overestimated for the middle section around 4.5 km downstream of the stream source. This may be due to vertical errors in the DEM used (Nikolakopoulos et al. 2006; Blanchard et al. 2010; Arefi and Reitnard 2011), limited historical information on metal mine production in the catchment, or a lack of accurate information relating to the location of a 20th century sediment retention dam, presumed to be responsible for the high accumulation of contaminated sediment observed in the field surveys around 4-5 km downstream in the river.

Despite these differences, general patterns of increase/decrease of contaminant sediment along the river channel are effectively captured by the model, and hotspots can be identified. Our results indicate good agreement between the field data in 2010 and the actual simulation runs. Validation was not possible in the Ampoi catchment, but, based on the good agreement found in the other two catchments, we have projected future contamination levels based on the current distribution of contaminated material in the catchment.

Historical simulation runs

Although historical data on mining waste production are available from 1791 and 1848 for the Ystwyth and Naracauli rivers, respectively, CAESAR runs were started 20 years earlier in order to provide a transitory time for sediment stabilization in the floodplain. In the Ystwyth, runs show immediate sedimentation and stabilization of waste material in the lower reaches of the floodplain. Only at the end of the 20th century did remobilization of waste material occur due to some extreme rainfall events that eroded the floodplain (Fig. 5b).

Historical records show production of mine waste reached peaks around 1860 and 1940 in the Ystwyth and Naracauli catchments, respectively (Fig. 6). Simulations in both catchments show that total Pb waste output at the outlets of both catchments respond differently to the forcing imposed by waste production in the point sources. Peak waste release in the Ystwyth outlet (c. 1930) occurred approximately 80 years after peak production (c. 1850), indicating a high sediment-associated retention capacity in the Ystwyth catchment. In contrast, waste release in the Naracauli peaked only 10 years after large mining production events, this is a consequence of the small size of the catchment, the low retention capacity and high erodibility of the catchment soils/sediments. The peaks in outlet waste release in the Ystwyth at the end of the 19th century correspond with maximum concentrations in the middle reaches of the catchment – particularly in the floodplain - reaching concentrations of 10000-40000 mg kg^{-1} in some locations, resulting from the high degree of coupling between the point source of Grogwynion (a mine in the middle of the catchment) and the river, which fed the narrow floodplain immediately downstream with hundreds of tonnes of waste material. These numbers are likely to be overestimates of the actual concentrations during those years, probably due to the high sensitivity to source location the model has in narrow reaches such as at Grogwynion.

In the Naracauli, high concentrations of pollutants start to develop at the end of the 20th century immediately downstream from the Brassey mine (top of the catchment). However, pollution is widespread across the catchment during the whole 20th century, and only during the last 30 years has the catchment released much of the floodplain polluted sediment into the sea. Whilst the spatial extent of the floodplain polluted areas did not change much following the onset of mining activities (Fig. 7), only recently in the upper reaches of the catchment have areas with high concentrations of sediments expanded. This slow expansion in the upper sections is due to the slow displacement of polluted material adjacent to most of the point sources, which are very weakly coupled to the floodplain.

Forecasting future catchment contamination

Future scenarios of soil and sediment contamination in the catchment are crucial in order to predict the consequences of both remediation and non-remediation scenarios. Simulations were started in 2020, because any potential remediation measures in the catchment are not likely to start immediately, and were continued up to 2100 to conform with standard forecasting dates for long-term scenarios such as those predicted by the IPCC (Intergovernmental Panel on Climate Change). In this study, we adopt a "best scenario" approach to remediation. Remediation, whether physical or biological, is assumed to be completely successful in the areas where it take place, both on point and diffuse sources. Therefore, in all those pixels (cells) rehabilitated we assume that a complete elimination of contaminated sediment has taken place. Interventions take place in mine spoils and tailings, as well as metal contaminants accumulated in floodplain sediments. Only the areas immediately adjacent to the river channel were left unremediated. We recognise that total 'point' and 'diffuse' remediation is probably unachievable in reality in the study catchments (although it might be feasible in the Naracauli), however, the scenario is tested in the model to ascertain the maximum benefit that potentially could be obtained from such an intervention. Both remediation and non-remediation simulations were run under the same stochastic rainfall series. While such action leads to underestimation of the potential variability associated to a full exploration of the inherent stochasticity due to weather patterns, it does allow direct comparisons to be established between remediation and non-remediation scenarios.

Remediation vs. non-remediation future projections in all three catchments show much higher polluted sediment releases in the Naracauli and Ampoi catchments, mostly during the first 15 years (Fig. 8). Particularly in the remediated scenarios, release of polluted sediment in the catchment oulets takes place within the first 10 years of the intervention. Both the number of events with intense outflow of polluted sediment and the actual volume of released contaminated sediment out of the catchment are actually higher in Naracauli and Ampoi, while the Ystwyth shows a more constant and slow release of Pb throughout the entire period of simulation (Fig. 8). In all catchments, non-remediation scenarios show patterns of slow and more continuous release of contaminated sediment. None of the three catchments seems to have reached stability in release patterns at the end of the simulation. However, point and diffuse remediation seems to be very effective, particularly in the Naracauli and Ampoi catchments, reaching practical stabilization by 2060, and very fast clean-up during the first 8 years following the instigation of remediation measures.

In the Naracauli, Pb levels in both the river and floodplain improve significantly in the remediation scenario (Fig 9). While projections under no remediation show certain reductions in the concentrations of polluted sediment (mostly upstream but also to a great degree in the lower part of the catchment), metal levels decrease because of reductions erosion of contaminated sediment and/or dilution by 'clean' sediment under remediation, and result in impressive reductions during the first half of the 80 year modelling run. However, given the steep gradients and high relief in the catchment, only the sediment concentration, and not the spatial extent of the sediment, is altered over time and as a result of the remediation intervention (Fig. 9a).

In the Ampoi River, there is currently a hotspot of high lead concentration (3500 mg kg^{-1}) around the tailing pond next to the town of Zlatna (Fig. 10). Future forecasts of Pb concentration levels without any remediation measures predict a large removal of sediment along the river in most areas (except the area surrounding the tailings pond) between 2020 and 2060. The situation, however, barely changes after 2060, indicating a great degree of sediment stabilization in the catchment. By contrast, in the remediated scenario the initial catchment response is to completely clean sections of the river above Zlatna town and the floodplain immediately upstream of Alba Iulia (at the mouth of the Ampoi). However in this downstream area concentrations increase to around 1200 mg kg^{-1} by 2100. This is due to the remediation strategy used: 'cleaning' the floodplain actually establishes a larger coupling between the river and the floodplain, allowing the system to accommodate and rapidly accumulate any contaminated sediment migrating downstream from the source areas. This results in very fast build-up of contamination in the floodplain. Due to the lack of validation up to the present day (2010), the predictions capabilities in the Ampoi are somewhat limited. However, given the very good results achieved in simulations in the other two catchments, we can be confident that, at least at coarse scales, and for the location of hotspots of polluted sediment, our results for the Ampoi follow overall the same pattern of dependence on geomorphologic conditions. Remediation does not contribute much to the cleaning of the catchment in the Ystwyth, probably due to the high retention capacity in the Ystwyth floodplain (Fig. 8). Removal of contaminated material is limited and only occurs in the upper reaches of the Ystwyth valley immediately downstream of the Cwmystwyth mines (Fig. 9b). In the floodplain, however, limited accumulation of contaminated sediment is expected to occur during the years up to 2100. Under remediation scenarios, however, there is a large reduction in concentrations of sediment at the beginning in the upper reaches, and strong wash-out of polluted material afterwards. This is mainly due to the enormous contribution to remediation produced by the remediation of the relatively large floodplain currently present.

A conceptual model for catchment remediation

Our results of modelling the effects of catchment-scale remediation show that remediation strategies can yield contrasting catchment-dependent rates and patterns of improvement in terms of in-channel sediments and floodplain quality. Reduced in-stream contaminant concentrations may immediately benefit the riverine ecosystems and the health of people living nearby, but in the long-term reduced metal delivery to the floodplain environment may be even more important for the health of people and livestock.

Remediation at point sources (e.g. individual spoil tips, tailings ponds, mine sites) will inevitably yield a local environmental benefit. However, to what extent can remediation at point sources yield an environmental benefit to downstream river channels and floodplains (i.e. reduce metal levels below a particular threshold value)? The trade-off between sediment concentration, which increases with erosion and water flow, and contaminant concentrations must be taken into consideration. For example, in a highly erosive catchment flushing out of contaminants from point sources will be high, but, in catchments with large upstream erodible areas, the volume of eroded sediment will be so large that overall contaminant concentrations (i.e. the ratio between contaminant and sediment) may be low, introducing potential benefits if a non-remediation strategy is taken.

Alternatively, catchment-scale benefits may only be gained by remediating both point and diffuse sources. Thus, the fundamental question that needs to be addressed is which remediation strategy is most effective in terms of cost and environmental benefit? There is no 'one-size-fits-all' answer to the question because the geomorphology and process regimes of each catchment also need to be considered. In some cases, as has been shown, remediation may be costly and produce little benefit.

Variable geomorphologic characteristics in different catchments lead us to propose a conceptual model of remediation, including the following elements and processes:

1. Point source number, location and volumetric output of contaminants.
2. Concentration of point source and diffuse source contaminants.
3. Diffuse source (e. g. floodplain) area, flood frequency and duration.

4. Suspended sediment transport rate and concentrations (highly dependent on the vegetation rainfall interception present).

5. Degree of lateral (and vertical) channel mobility.

6. Catchment size, drainage density and/or climate/precipitation regime.

The variability of geomorphologic conditions in the three catchments studied highlights the remediation conundrum we must deal with. The modelled catchments encompass the following:

1. Ystwyth: Moderate gradients, maritime temperate climate with constant rainfall throughout the year, alternating laterally mobile/stable reaches and variable floodplain storage capacity.

2. Naracauli: Steep gradient, Mediterranean climate with extreme rainfall seasonality and individual intensive rainfall events, limited lateral mobility and floodplain storage capacity.

3. Ampoi: Relatively steep gradient, continental temperate climate with some degree of seasonality with a partially dry season in the autumn, limited lateral mobility but extensive floodplain storage.

With respect to these study catchments, the three most significant variables in terms of catchment-scale improvement potential would appear to be: *contaminant source coupling*, the *ratio of contaminated to non-contaminated sediment delivery*, and the *frequency of sediment transport events*. Figure 11 plots these variables in the form of a ternary diagram and superimposes the present ‘location’ of the three study catchments in the phase space. The green arrow represents the trajectory for improving catchment sediment quality either through natural ‘self-cleaning’ processes (e.g. sediment dilution) and/or active intervention via point/diffuse remediation strategies. Based on field data, Figure 11 suggests that the Naracauli catchment has the most significant present-day management problem, however, the CAESAR/TRACER model outputs indicate that it can be successfully remediated, likely through a combination of point and diffuse source interventions. The larger Ystwyth and Ampoi catchments have less severe contamination issues. However, the model scenarios studied here indicate that remediation interventions would yield a much greater environmental dividend on the Ampoi than on the Ystwyth (Fig 8).

Conclusions

The vast majority of sediment deposition events in catchments take place usually in areas far downstream from the steep gradients typical in upland areas. Since mining activities are in most cases located in those upland areas, sediment contamination produced from waste material will naturally tend to accumulate in the floodplains and lower parts of many of those catchments. This situation is present in the three catchments studied here.

Consequently, any remediation measure at the local scale in the point sources should be complemented by other, more extensive interventions in the floodplains. These contaminated floodplain sediments are a legacy of metal mining in the catchment and are of concern for two reasons. First, often these floodplain soils are being used for grazing livestock and for growing fruit and vegetables; if these elements are bioavailable there is then a risk that contaminant metals may enter the food chain. Second, these contaminated floodplain sediments are a diffuse store of contaminants and could be re-introduced into the river (such as the Ampoi) through bank erosion and surface wash processes.

The three contrasting catchments were selected in order to contrast the different hydrological and geomorphological regimes and review the potential effects of remediation measures associated with them. More realistic predictions would likely involve realisations of hundreds of simulation runs per scenario and catchment, given the stochastic nature of the rainfall model (see Coulthard et al. 2012). The probabilistic predictions resulting from this would then more clearly identify the confidence levels of our approach. However, observations from some trial runs indicate that the model is very robust to the size and timing of flood events, and more contingent on the floodplain morphology, which directly influences the three variables in our conceptual model.

While floodplain geomorphology greatly influences the distribution of contaminated hotspots in the catchment, it also contributes to the timing of contaminant release from the outlet. The striking differences between the Ystwyth and Naracauli rivers, both in rainfall frequency and intensity, and in geomorphological attributes produce high retention capacity of contaminants in the first and low in the second. This differential in retention translates into high delays in flush-outs in the Ystwyth and very rapid responses to contaminant release from the mines in Naracauli (Fig. 7). Thus, both spatial and temporal patterns of formation of hotspots are consequences of geomorphology, and provide differential potential recipes for remediation in terms of long- versus short-term management.

As expected, the effects of remediation are different in the three catchments, and sometimes may even be counterintuitive. For example, the Ampoi is a river with acceptable Pb concentrations levels in the channel sediment and apparently safe previous interventions with the construction of tailing dams. If this is a permanent improvement in channel sediment quality due to these dams then it suggests that ‘active’ on-site management, coupled with contaminated sediment ‘passive’ flushing by flood flows, may be an effective means of remediating mining impacted river systems in similar locations.

Our conceptual model, supported by our realistic simulations, intends to establish general guidelines following the topographic, climatic, and historical mining conditions of every catchment. Remediation measures will be in general more effective in those areas with low contaminant-source coupling, given by low topographic gradients, actual artificial barriers, or climates lacking intense rainfall events. Geomorphological and climatic conditions will as well determine the mine waste to clean sediment ratio (which will make remediation more difficult when its value is high). Finally, the frequency of sediment transport events, while potentially increasing the erosion and sedimentation of contaminants from the point sources, will likely contribute to dilute these with the clean sediments deposited from non-contaminated areas, facilitating the actions of human remediation interventions.

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Figure captions

Figure 1. Location map within Europe of the three study catchments.

Figure 2. Modules and inputs necessary to run CAESAR/TRACER. A new module for realistic rainfall generation has been added in this study (red frame) describing the procedures to follow for the creation of synthetic, hourly rainfall data series.

Figure 3. Examples of stochastic rainfall generation runs for the three sites studied. Yearly precipitation for the a) Ystwyth, b) Naracauli catchment, and c) Ampoi catchments. Upper panels: solid circles are cumulative numbers of storm, red lines are cell events. Middle panels are rainfall intensities (mm/h). Lower panels depict the monthly accumulation of rainfall in mm.

Figure 4. Lead concentrations (XRF values) in river sediment of the a) Ystwyth, b) Naracauli and c) Ampoi rivers.

Figure 5. a) Simulated and field data for the fine fraction of lead in the Ystwyth catchment profile. b) Historical development of polluted areas in the Ystwyth catchment and close view of the main floodplain in the middle reaches of the catchment. The easternmost polluted area is the hotspot located just downstream from Cwmystwyth village. Observe the expansion of the polluted floodplain area during the last 80 years.

Figure 6. Recorded historical waste production (black line) and simulated lead waste output (in red) at the outletss of the Ystwyth and Naracauli catchments.

Figure 7. Historical development of polluted areas in the Naracauli catchment and close view of the upper catchment. Observe the development of highly polluted areas (red) during the last 80 years in all areas (the Gennamari mining complex being the most affected) and a hotspot in the floodplain downstream from the main Brassey mill.

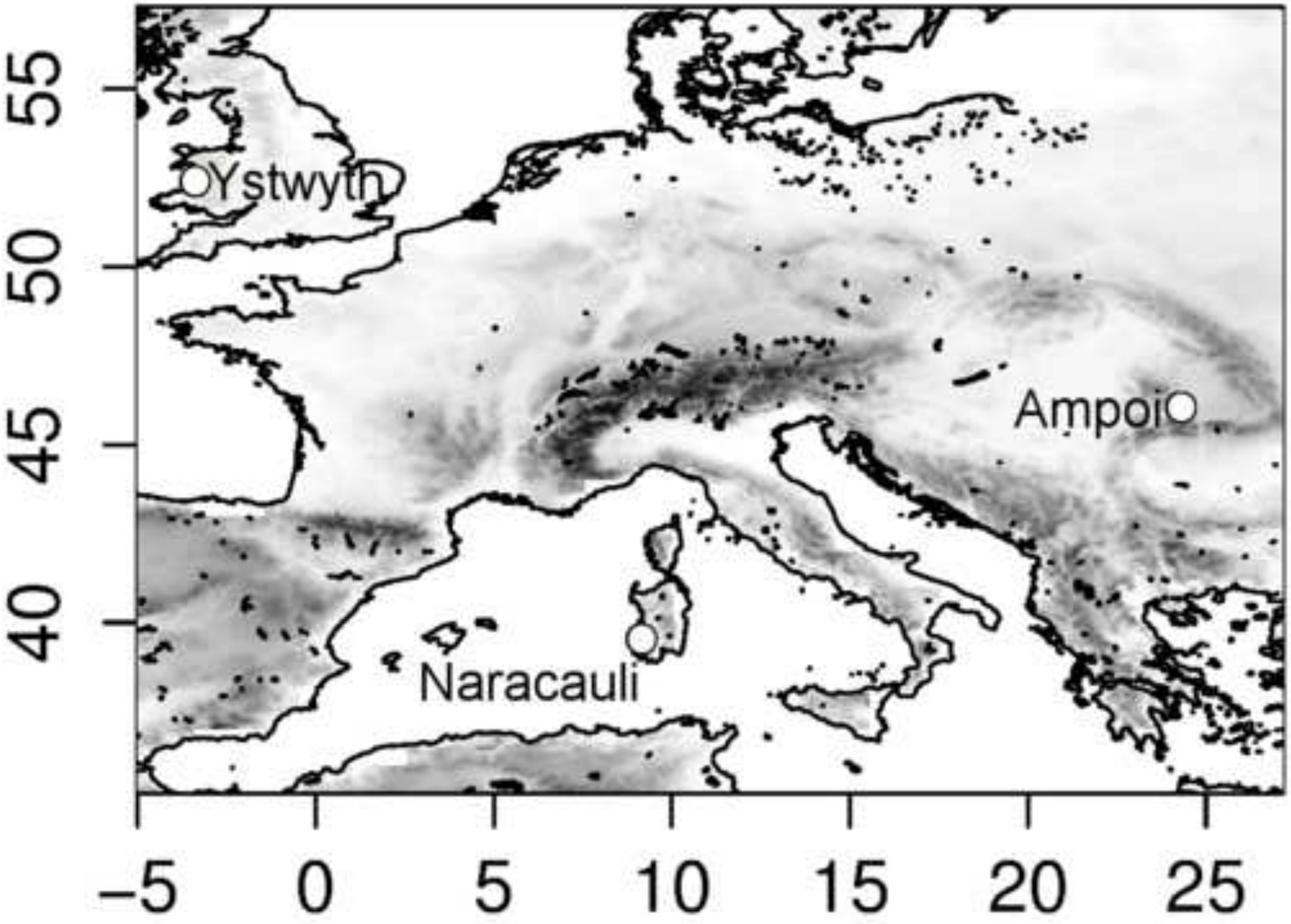
Figure 8. Simulated cumulative release of contaminated sediment in the three catchments studied and their respective projection scenarios. Red lines: No remediation. Green lines: Point and diffuse remediation.

Figure 9. Future development of polluted areas in two catchments. a) Naracauli: enlarged view, similar in both remediation and non-remediation scenarios. Upper panel: Lower sections of the catchment. Lower panel: Upper sections of the catchment. Observe the continuous cleaning of the upper sections, mostly around the Gennamari mining complex. b) Ystwyth: Upper panel: non-remediation. Lower panel: Remediation. Observe a highly polluted area (red dot) upstream and the reduction of the floodplain surface under the remediation scenario.

Figure 10. Future development of polluted areas in the Ampoi. a) Non-remediation scenario in the whole catchment (upper pannel) and close view of the main sources in the middle of the catchment (lower panel). The brown areas in the upper close view are the Zlatna town tailing pond and the Iazul tailing pond. b) Remediation scenario in the catchment (upper panel) and close view of the floodplain near Alba Iulia (lower panel). The remediated floodplain rapidly builds up again due to sediment recirculation.

Figure 11. A conceptual model for catchment remediation. The green arrow represents the optimal direction for a catchment-scale remediation strategy. The ellipses show the conceptual locations of the three catchments studied (Y: Ystwyth, N: Naracauli; A: Ampoi) are shown.

Figure 1



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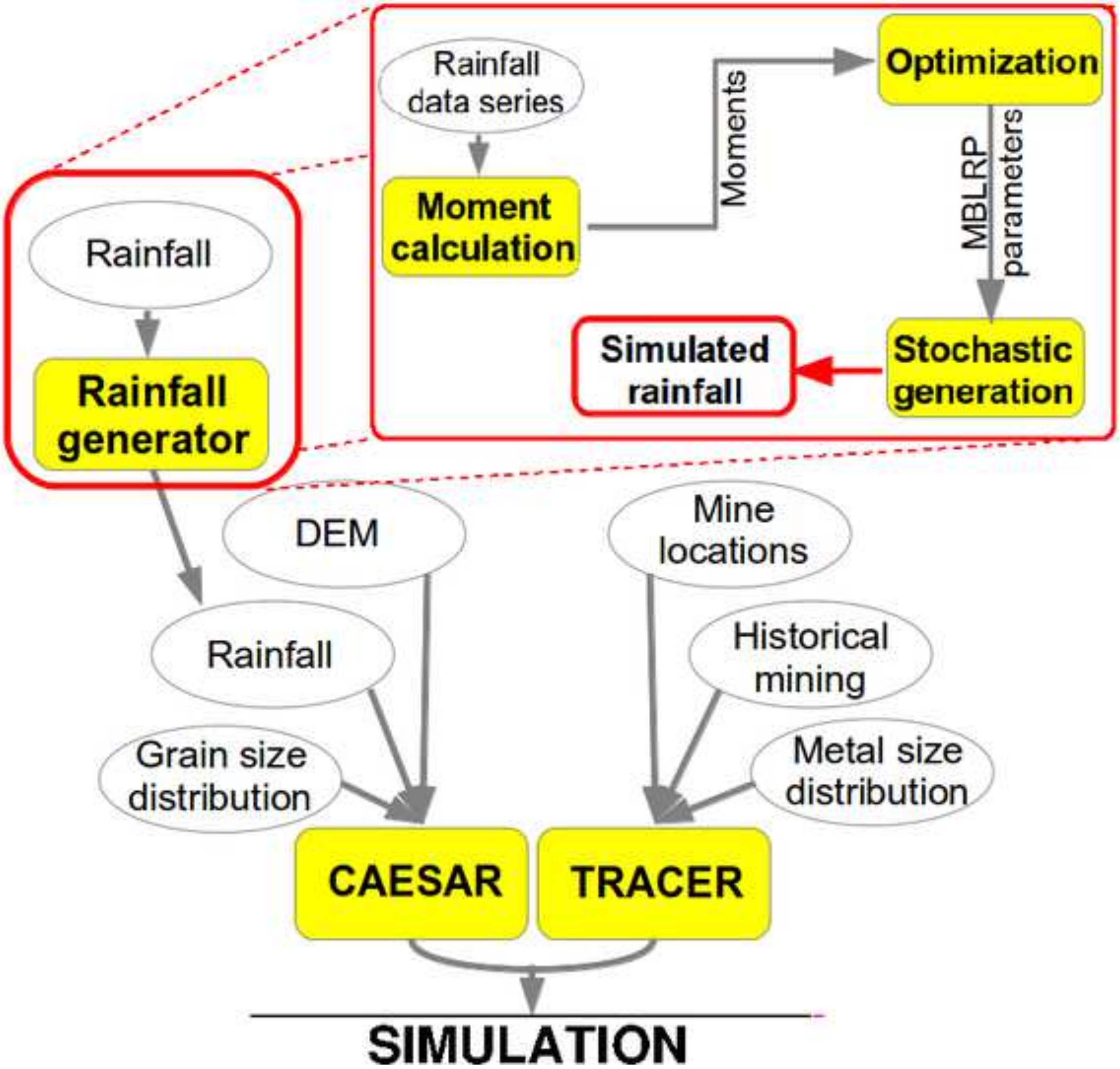
Rainfall data series
Moment calculation
Moments
Optimization
MBLRP parameters
Simulated rainfall
Stochastic generation
Rainfall
Rainfall generator
DEM
Mine locations
Rainfall
Historical mining
Grain size distribution
Metal size distribution
CAESAR
TRACER
SIMULATION

Figure 3

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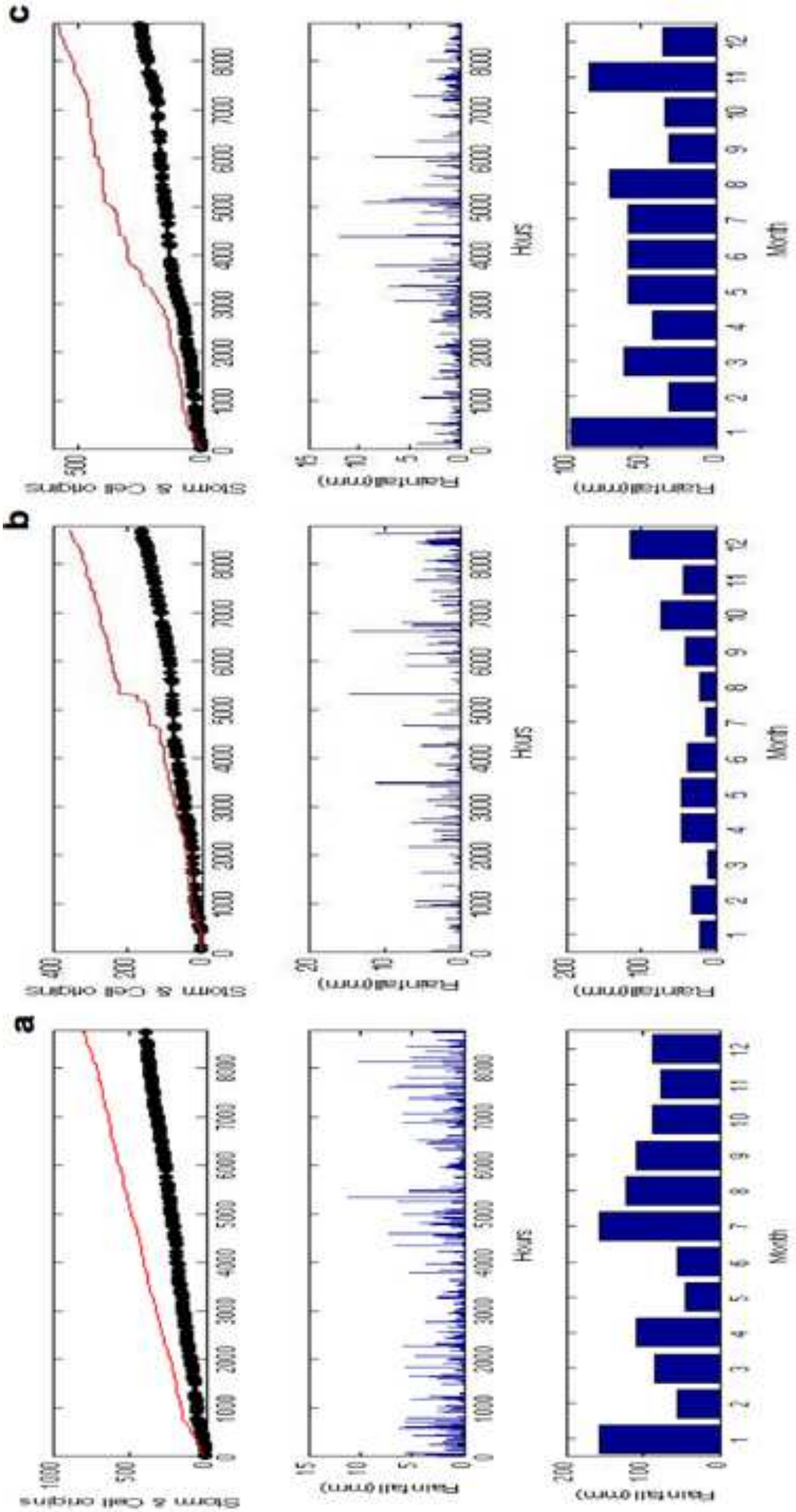

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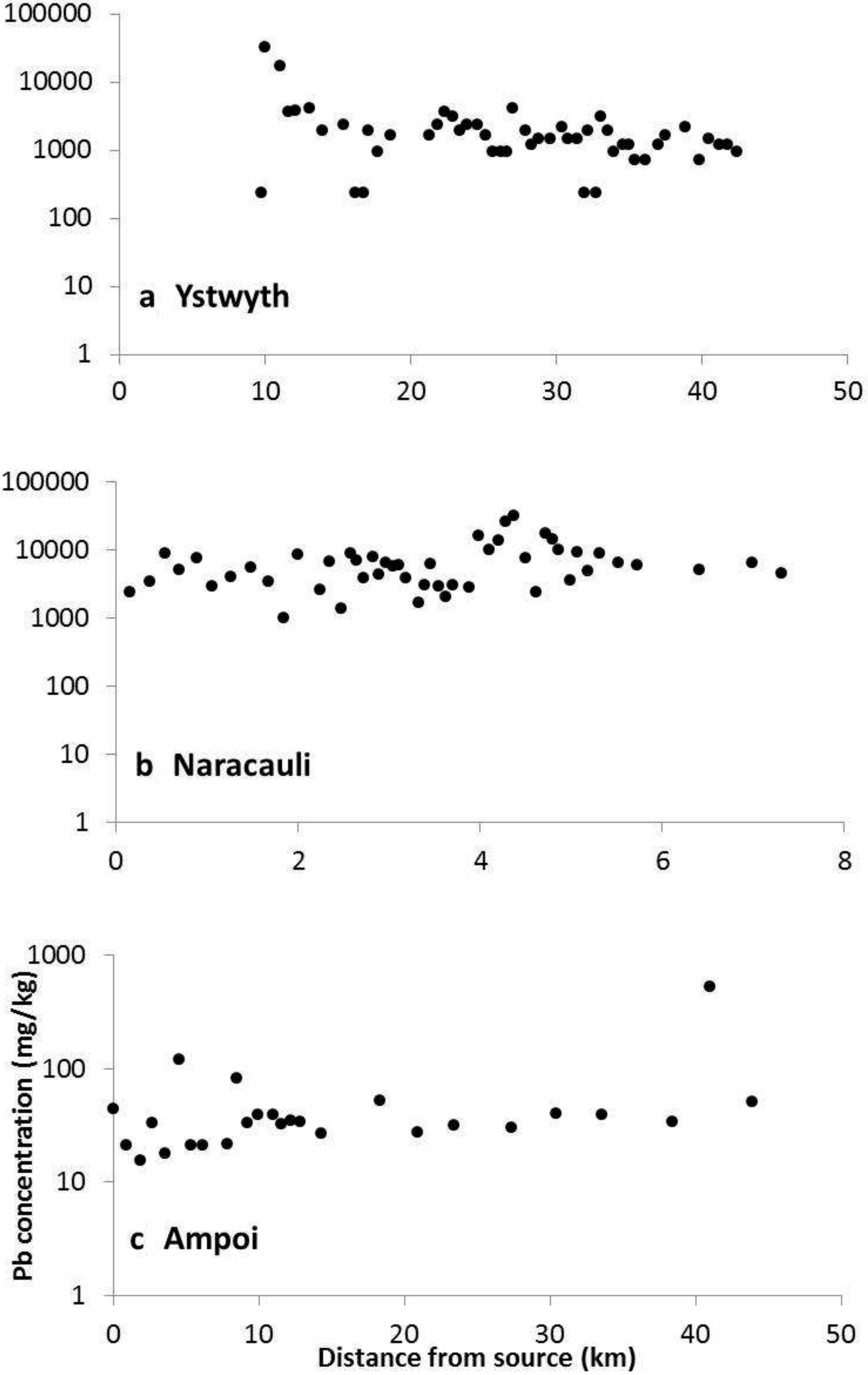
a Ystwyth
b Naracauli
c Ampoi
Pb concentration (mg/kg)
Distance from source (km)
100000
10000
1000
100
10
1
0
10
20
30
40
50
2
4
6
8

Figure 5

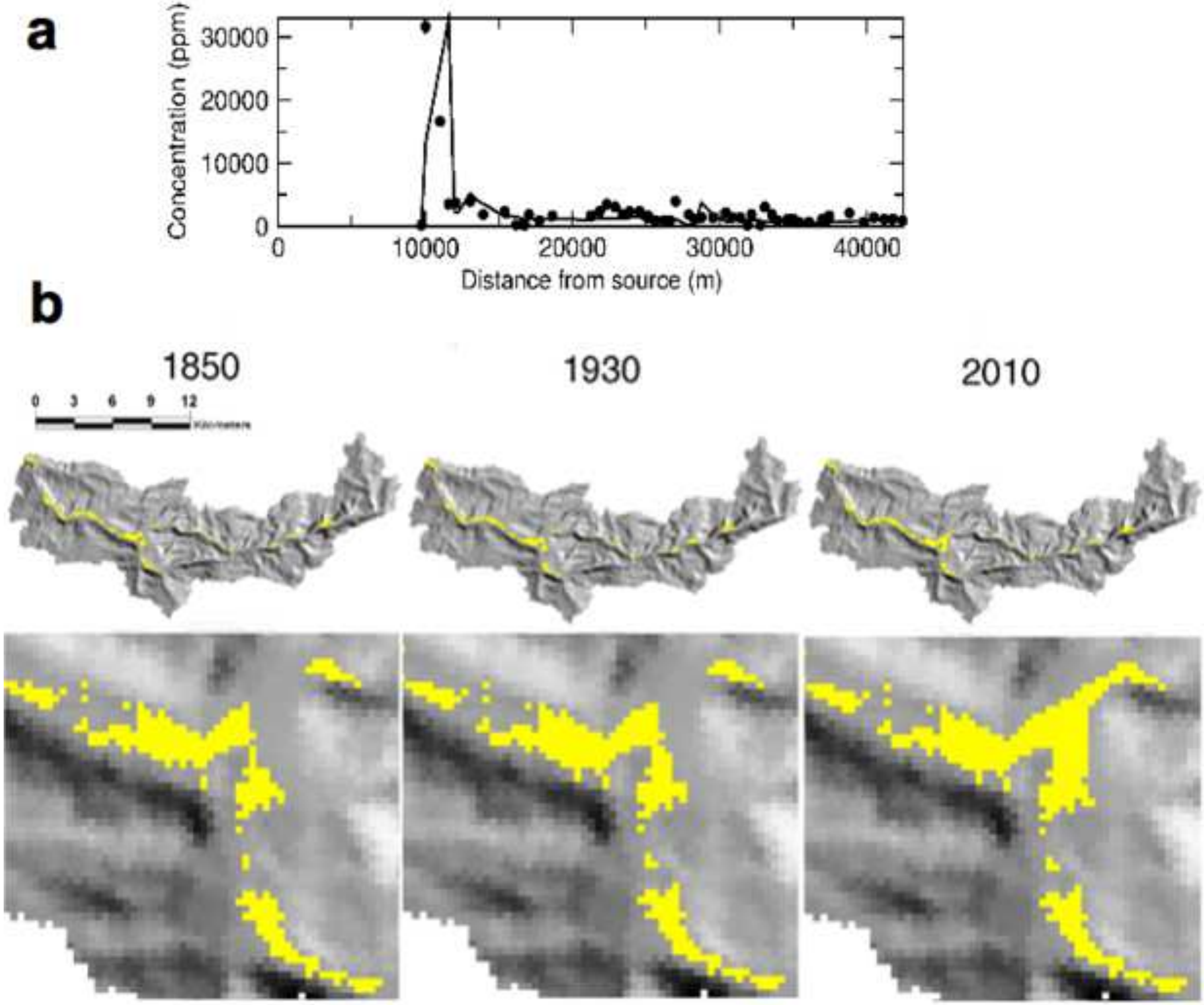
a
Concentration (ppm)
30000
20000
10000
0
0
10000
20000
30000
40000
Distance from source (m)
b
1850
1930
2010
0
3
6
9
12
Kilometers

Figure 6
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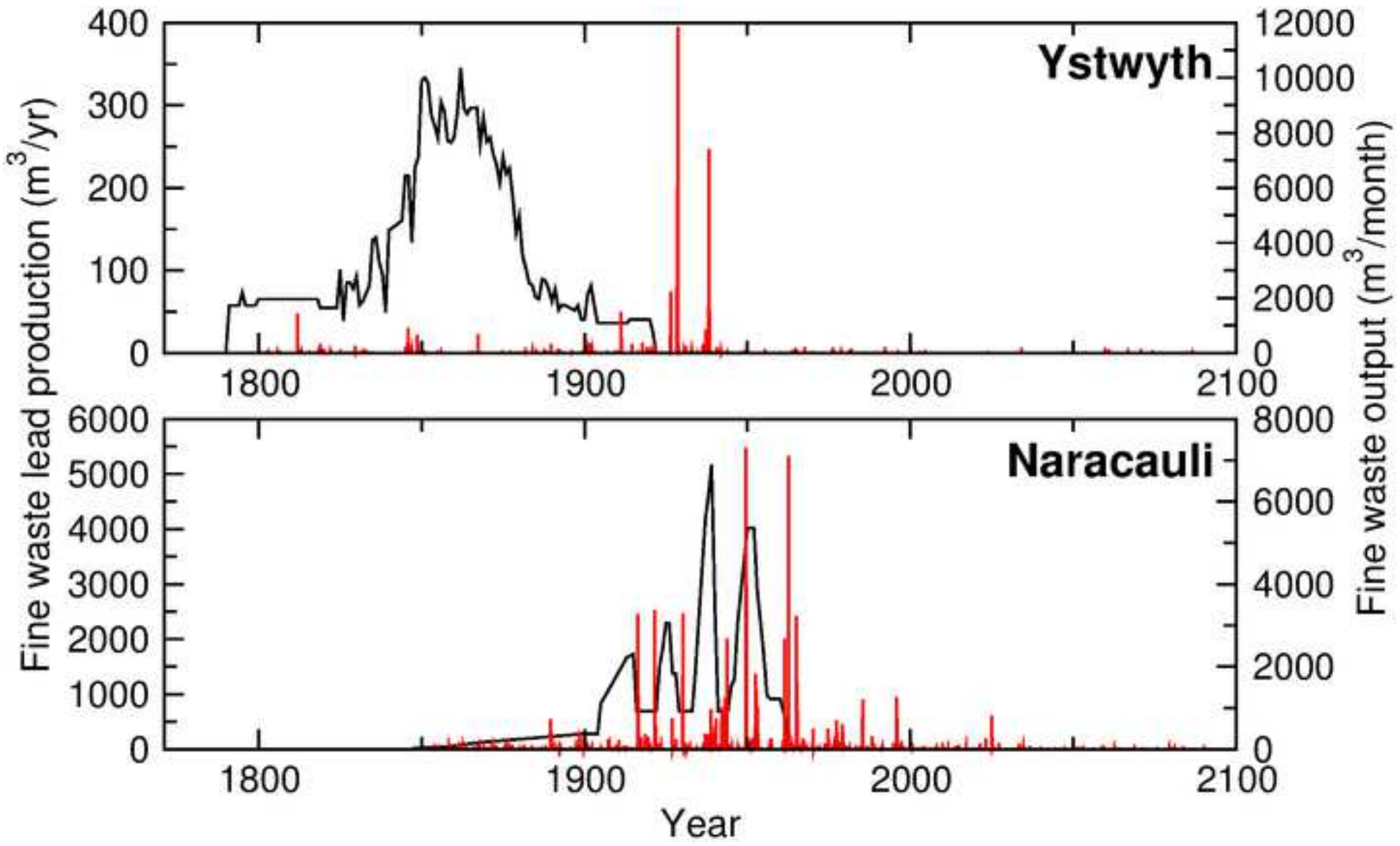

Figure 7

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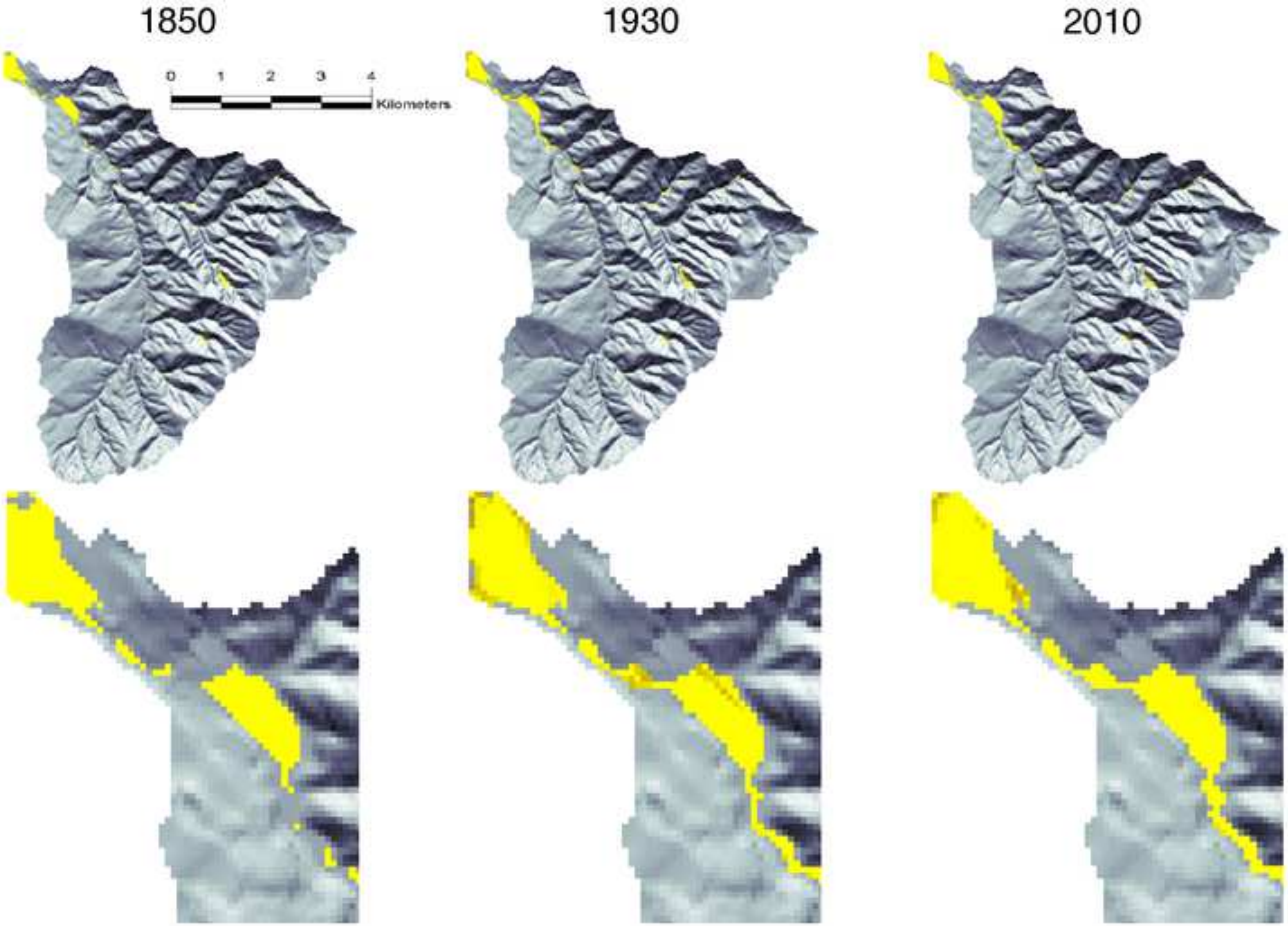

Figure 8

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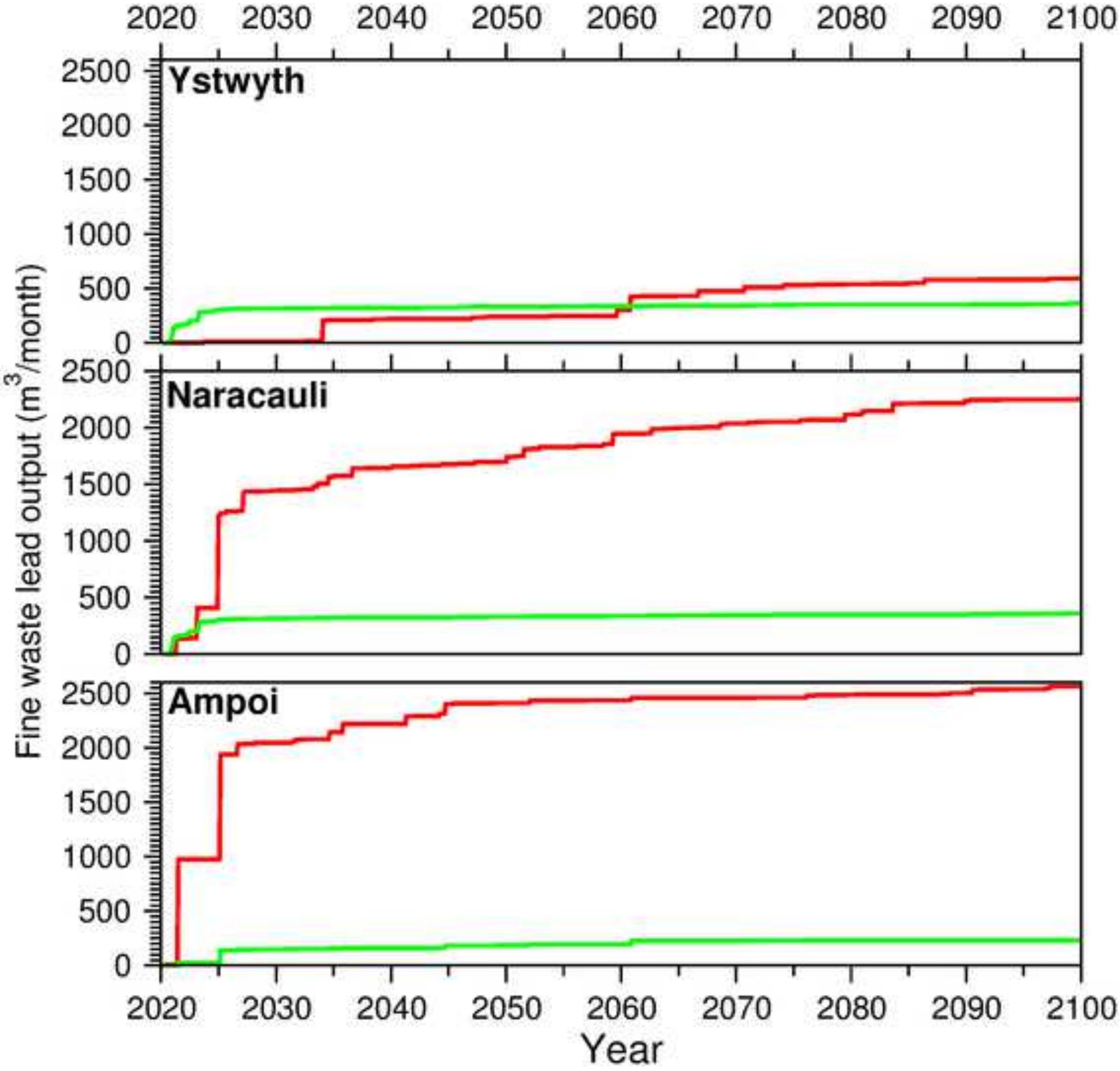


Figure 9

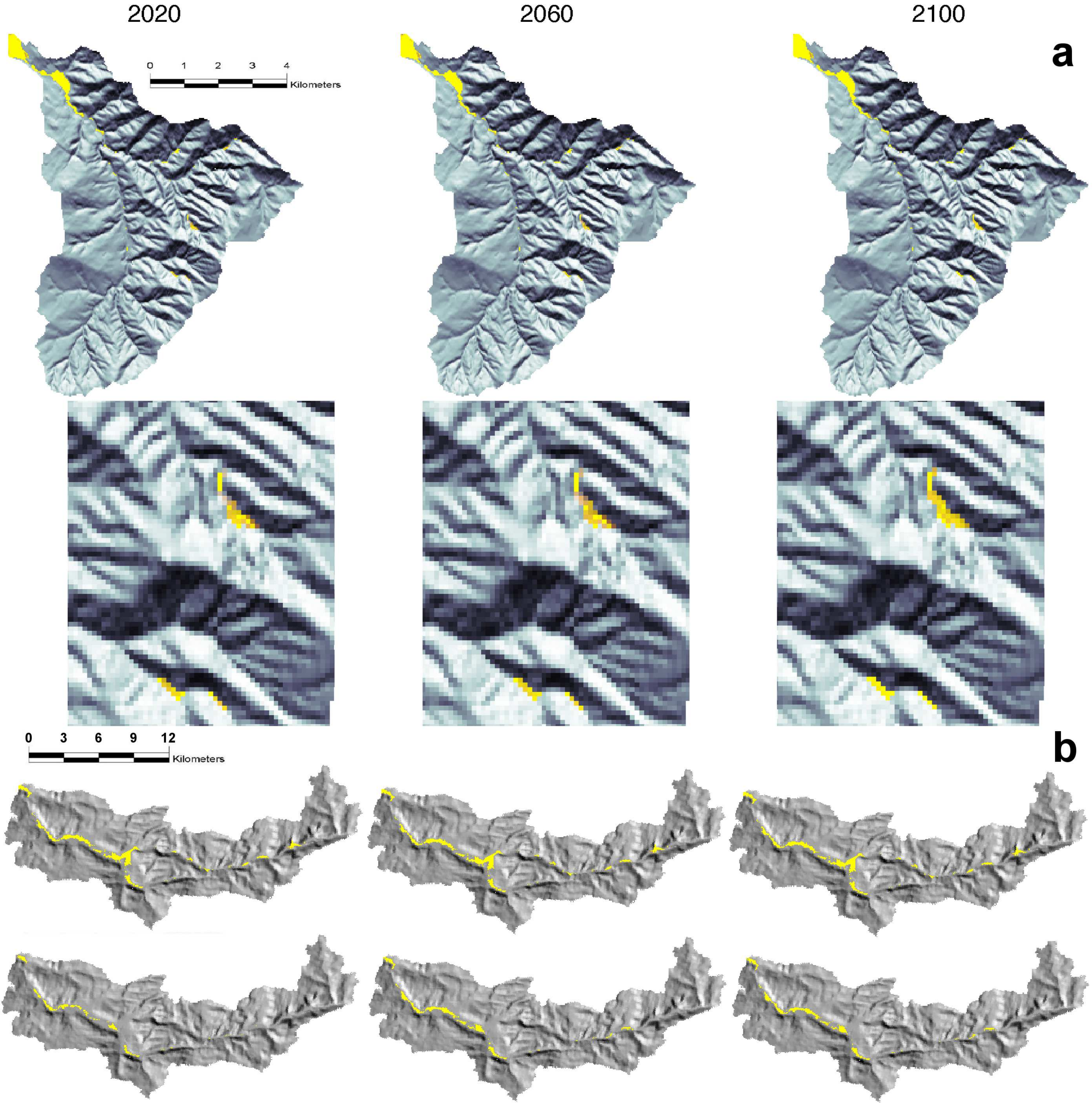


Figure 10

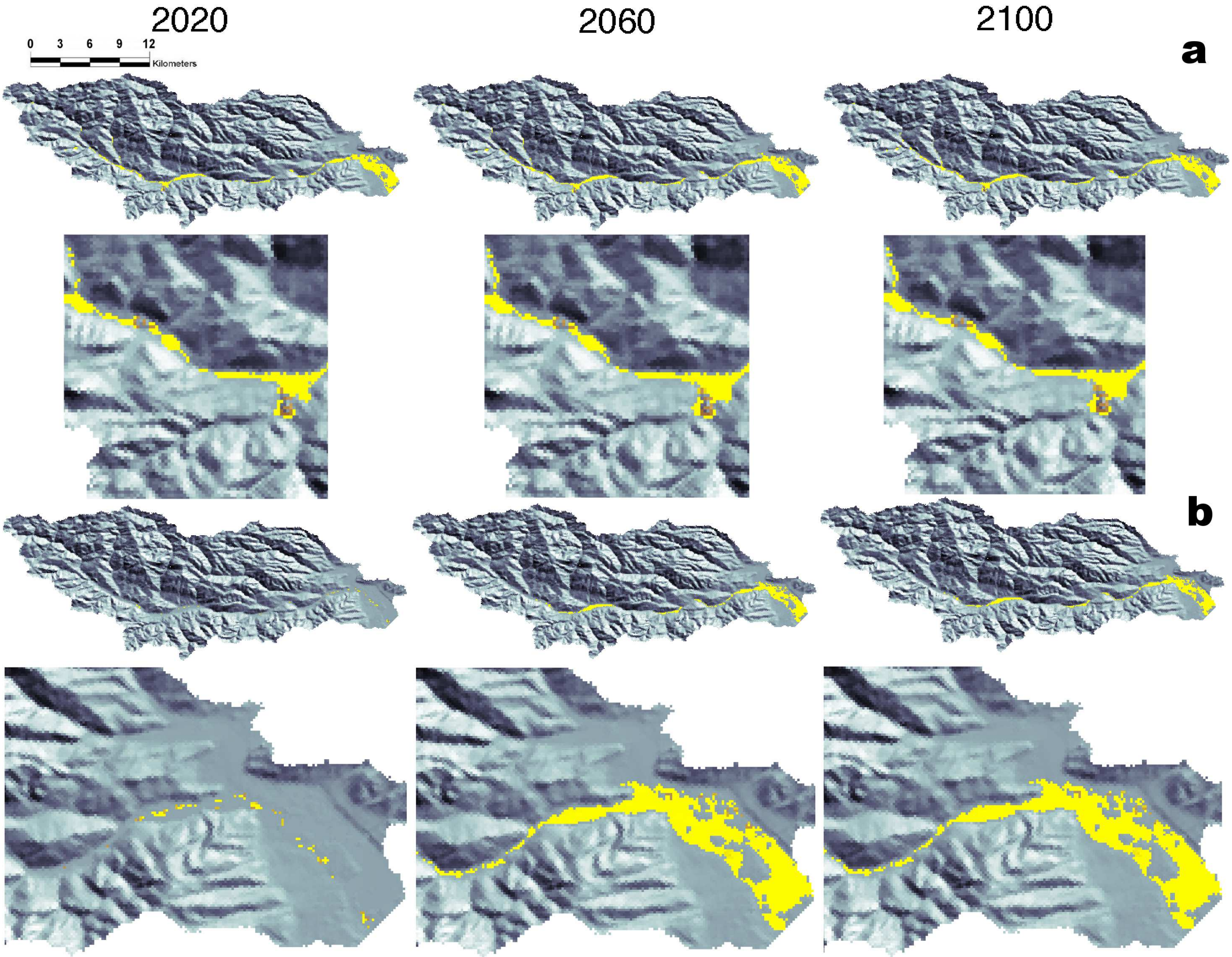
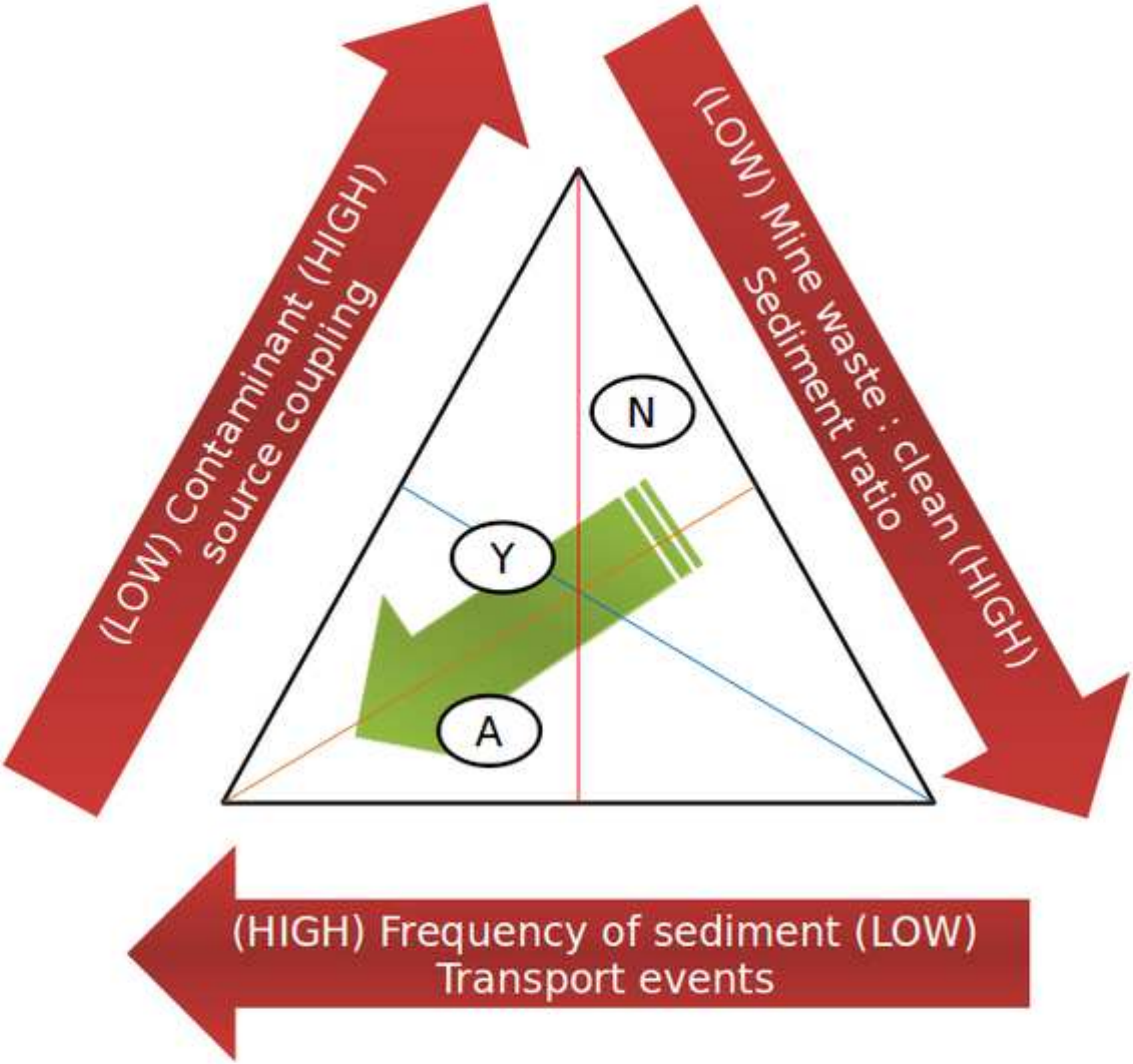


Figure 11


(LOW) Contaminant (HIGH)
source coupling
(LOW) Mine waste : clean (HIGH)
Sediment ratio
(HIGH) Frequency of sediment (LOW)
Transport events
N
Y
A