



Dissolved trace element concentrations and fluxes in the Irrawaddy, Salween, Sittaung and Kaladan Rivers

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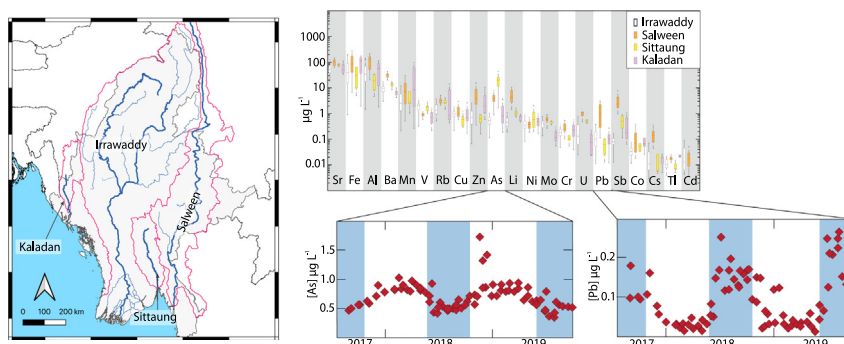
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HIGHLIGHTS

- The Irrawaddy and Salween are chronically understudied globally important rivers.
- Extensive sampling campaign used to determine trace element fluxes in these rivers.
- Irrawaddy-Salween supply ~1–17 % of global dissolved riverine trace elements fluxes.
- Most elements are unperturbed by anthropogenic activities in the studied rivers.
- Exceptions include As in the Sittaung River which is a concern for human health.

GRAPHICAL ABSTRACT



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ABSTRACT

The Irrawaddy (Ayeyarwaddy) and Salween (Thanlwin) globally rank among the largest rivers for supplying dissolved and particulate material to the ocean. Along with the Sittaung and Kaladan rivers they have societal importance to Myanmar in terms water sources and food production. Despite their importance for global biogeochemical cycles and the ~50 million people who live in their catchments, the chemistry of these rivers is poorly known. This study presents a comprehensive survey of dissolved (<0.22 µm) trace element concentrations (Sr, Fe, Al, Ba, Mn, V, Rb, Cu, Zn, As, Li, Ni, Mo, Cr, U, Pb, Sb, Co, Cs, Tl and Cd) at 38 locations within these river catchments, spanning a period of 2 years. The results highlight the global importance of the Irrawaddy and Salween rivers for trace element global biogeochemical cycles; contributing between 1 and 17 % of global dissolved riverine fluxes to the land-ocean interface for the studied elements. Area normalized dissolved fluxes in these catchments are ~2 to 10 times higher than global average values for most elements, consistent with high rates of chemical weathering. In general, anthropogenic activities have yet to significantly perturb dissolved trace element fluxes in these river systems. The presented dataset should therefore serve as a useful 'natural' baseline, against which future perturbations driven by climate change and/or the development of Myanmar's mining industry could be assessed. Exceptions to this include As in the Sittaung River and Sb, Zn,

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Pb and As in the Salween River, which may already be significantly impacted by anthropogenic inputs. The former represents a water quality issue of concern for public health, and so constraining the exact sources of As in the Sittaung River should be considered a priority for future research.

1. Introduction

The Irrawaddy (Ayeeyarwaddy), Salween (Thanlwin), Sittaung and Kaladan rivers play a central societal role in Myanmar by providing important water sources and fisheries, and by supplying fertile soils to major agricultural regions (Kattelus et al., 2014; Taft and Evers, 2016; Ketelsen et al., 2017; Johnston et al., 2017). The Irrawaddy and Salween are also of importance in terms of global biogeochemical cycles. They globally rank 12th and 26th in terms of freshwater discharge, and likely both rank within the top 10 rivers globally in terms of fluxes of both dissolved and particulate material to the ocean (Robinson et al., 2007; Bird et al., 2008; Furuichi et al., 2009; Milliman and Farnsworth, 2011; Manaka et al., 2015; Chapman et al., 2015; Baronas et al., 2020). Despite the importance of these rivers, both to the ~50 million people who live in their catchments and for constraining global biogeochemical cycles, they have been the subject of relatively few scientific studies. Recent studies have greatly improved constraints on the fluxes of major dissolved elements (Chapman et al., 2015) and particulate material (Baronas et al., 2020) of these rivers. However, there remains a lack of data on dissolved trace element concentrations and fluxes within these river systems. Addressing this knowledge gap is vital for assessing water quality in these catchments; many of these trace elements are potentially toxic with impacts on riparian biodiversity (e.g. Johnson et al., 2017), and to human health via drinking water, and bioaccumulation into crops and fish (e.g. Satarug et al., 2003; Berzas Nevado et al., 2010; Tuli et al., 2010).

Dissolved riverine trace element concentrations/fluxes are controlled by their supply, mainly via chemical weathering in addition to anthropogenic processes (e.g. mining, industrial and municipal waste water discharge), versus their removal to particulate phases controlled by the riverine dissolved (e.g. pH, Eh, and ligand concentrations) and particulate (e.g. reactive mineral surfaces) chemistry. Determining the dissolved trace element concentrations and fluxes in these river systems is of timely importance, given the potential for perturbations caused by climate change and economic developments in the coming decades. Further development of the mining industry in Myanmar in the near future represents a particular concern for perturbing dissolved trace element fluxes in these river systems. Myanmar is rich in a variety of metal ore deposits including Sn-W, rare earth elements, Cu, Au, Pb-Zn-Ag and Sb (Gardiner et al., 2014; Kyi Htun, 2014; Khin Zaw, 2017). At present, mining contributes only a small fraction of the economy of Myanmar, but has large potential to expand in the future (Gardiner et al., 2014; Kyi Htun, 2014; Khin Zaw, 2017). In turn this could significantly alter the mobilization of trace elements in these river catchments, with negative consequences for water quality. Land disturbance by mining activities has increased in these catchments in recent decades (LaJeunesse et al., 2016; Bhagwat et al., 2017) and there is already concern over water quality in parts of these river catchments due to mining related trace element pollution (MacTavish et al., 2018; Kawakami et al., 2019; Kyaw Zay Ya et al., 2020; Shrestha et al., 2020). A lack of data in these river catchments however currently hinders a rigorous assessment of such concerns.

This study presents a comprehensive survey of river water dissolved trace element concentrations (Sr, Fe, Al, Ba, Mn, V, Rb, Cu, Zn, As, Li, Ni, Mo, Cr, U, Pb, Sb, Co, Cs, Tl and Cd) in the Irrawaddy, Salween, Sittaung and Kaladan river catchments, comprising of 550 samples. Some other trace elements are not reported, including rare earth elements that were typically below detection, and Hg that requires specialist collection and measurement approaches. These elements are also of interest for mining and toxicity assessment, but fall outside the scope of this study. These data are used to assess (1) the importance of these rivers in terms of trace element global biogeochemical cycles, (2) the quality of these water sources

with respect to trace elements, (3) the current role of anthropogenic activities for perturbing trace elements in these river systems, and (4) to provide a baseline to assess future perturbations to trace element fluxes in these river systems.

2. Materials and methods

2.1. Study area and samples

Data are presented for time-series of river water samples collected at 18 locations within these catchments, at a sampling frequency of once or twice a month, spanning 2 years and the full range of river discharge stages (Fig. 1, Supplementary Table 1). In addition, river water samples were taken twice yearly at 20 locations close to mining and industrial activities that are of concern in terms of trace element pollution (Fig. 2).

2.1.1. Irrawaddy River catchment

The majority of the Irrawaddy River catchment (413,710 km²) lies within Myanmar, and has a population of 39.5 million people (Ketelsen et al., 2017) (Fig. 1). The head of the Irrawaddy is at the confluence of the rivers Mali Kha and Nmali Kha about 50 km north of the town of Myitkyina. It extends ~2000 km south, draining into the Andaman Sea via a network of distributaries of the Irrawaddy River delta. It ranks 12th globally in terms of water discharge to the ocean (Milliman and Farnsworth, 2011), with an annual average water discharge of $379 \pm 47 \text{ km}^3 \text{ yr}^{-1}$ (Furuichi et al., 2009). The Chindwin River, the largest tributary to the Irrawaddy River, supplies about 50 % of this total water discharge (Chapman et al., 2015). Seasonal changes in Irrawaddy discharge are predominantly driven by the southwest Asian monsoon, with mean monthly discharge at Pyay, just upstream from the delta, varying from about 5 to 20 km³/month during the dry season of November to May, to 10 to 90 km³/month during the monsoon season of May to October (Furuichi et al., 2009).

The headwaters of the Irrawaddy River originate in the mountains of the eastern Himalayan syntaxis in the north of the catchment, with a maximum elevation of 5881 m (Taft and Evers, 2016). The western and eastern margins of the catchment also feature mountainous regions of the Indo-Myanmar ranges and Shan plateau respectively. These mountainous regions are sparsely populated, largely covered by relatively pristine forests (Salmivaara et al., 2013; Bhagwat et al., 2017). The majority of the catchment's population resides in the low-lying central valley, and delta regions of the river catchment, where land use is dominated by agriculture (Salmivaara et al., 2013). These regions include populous cities, including the largest city in Myanmar – Yangon – with a population of ~5 million people. The central valley receives relatively little precipitation due to its location in the rain shadow of the surrounding mountainous areas (Win Win Zin and Rutten, 2017). Agriculture in this region is therefore highly reliant on irrigation (Kattelus et al., 2014; Taft and Evers, 2016; Ketelsen et al., 2017). Irrigation consequently represents the dominant water use in the Irrawaddy catchment (accounting for ~90 %), followed by domestic use (~10 %), with rivers being a key source of this water (Kattelus et al., 2014; Taft and Evers, 2016).

The distribution and type of known ore deposits in the catchment are shown in Fig. 1 (Khin Zaw, 2017; Khin Zaw et al., 2017; Ye Myint Swe et al., 2017; Than Htun et al., 2017a, 2017b and Toe Aung Kyaw, 2017). Many of these deposits are being actively mined, often using small-scale/artisanal techniques (Kyi Htun, 2014; Aung Kyin, 2014). They include Au, Cu and Pb-Zn-Ag deposits associated with the Mogok metamorphic belt and sedimentary rocks of the Shan plateau that underlie the upper Irrawaddy catchment (north of Mandalay), and the

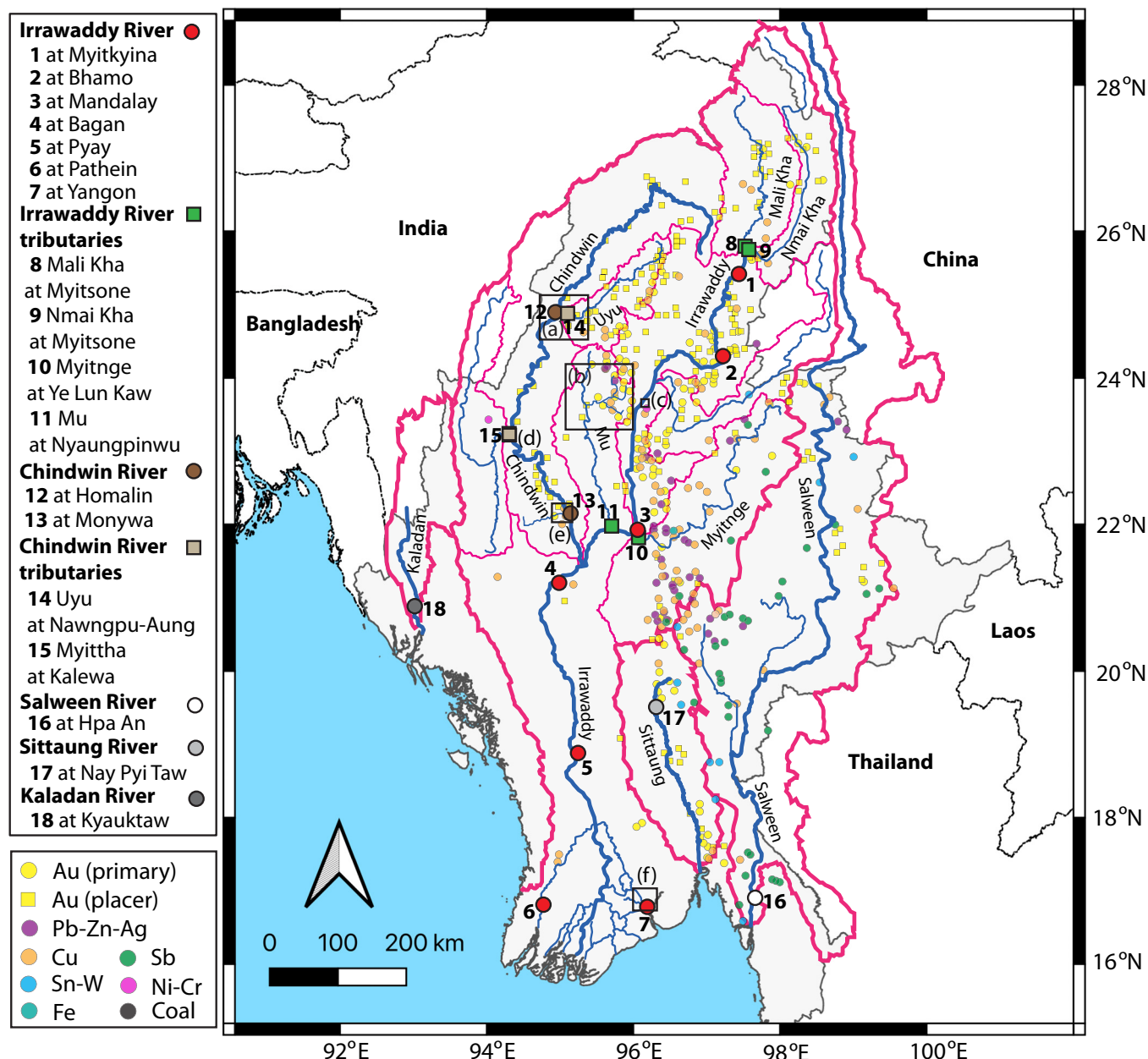


Fig. 1. Locations of river water time-series sampling stations (numbers) and mineral deposits within the studied river catchments. Light grey shaded region denotes Myanmar. Black squares labeled (a) to (f) show areas of maps shown in corresponding panels of Fig. 2. The boundaries for the Irrawaddy, Salween, Sittaung and Kaladan river catchments are denoted by thick pink lines. Sub-catchment boundaries within the Irrawaddy basin denoted by thin pink lines. International boundaries are denoted by dashed black lines. River channel and catchment boundaries sourced from HydroSHEDS (Lehner et al., 2008) and HydroBASINS (Lehner and Grill, 2013). Locations of mineral deposits are from Khin Zaw (2017), Khin Zaw et al. (2017), Ye Myint Swe et al. (2017), Than Htun et al. (2017a, 2017b) and Toe Aung Kyaw (2017).

catchments of the Myittha, Mali and Nmai Rivers (Gardiner et al., 2014). Additional clusters of Au and Cu deposits are associated with the Wundwin-Popa volcanic arc are found in the catchments of the rivers Mu and Uyu, tributaries of the Irrawaddy and Chindwin Rivers respectively (Gardiner et al., 2014).

Time-series river water samples were collected for this study at 15 stations within the Irrawaddy River catchment (Fig. 1, Supplementary Table 1). These include 5 stations on the main channel of the Irrawaddy River (stations 1 to 5), 2 on delta distributaries (stations 6 and 7), and 2 stations on the headwater rivers; the Mali Kha (station 8) and Nmai Kha (station 9). The rivers Mu and Myittha, major tributaries to the Irrawaddy River, were sampled at stations 10 and 11 respectively. The Chindwin River, the largest tributary to the Irrawaddy River, was sampled at two stations (12 and 13), with 2 additional stations that sampled

the major Chindwin River tributaries, the rivers Uyu (station 14) and Myittha (station 15).

In addition, samples were collected from 20 locations close to mining and industrial activities at a frequency of twice a year within the Irrawaddy River catchment (Fig. 2, Supplementary Table 1). Eight of these sampling locations are in the lower Uyu and upper Mu River catchments, areas that are heavily impacted by artisanal Au mining operations (Bhagwat et al., 2017, Shrestha et al., 2020, Aung Zaw Tun et al., 2020). Two of these sampling locations are on the Myittha River, close to coal mining activities (Khin Zaw, 2017). Four are situated on Yamar Creek and the Chindwin River, proximal to the large Sabetaung and Letadaung Cu mines, close to the city of Mawlaik (Khin Zaw et al., 2017). Six of these samples are on the Irrawaddy River, and wastewater drainage canals/ponds close to the ore refinery facility for the Tagaung Taung Ni-Cr deposit (Khin Zaw, 2017).

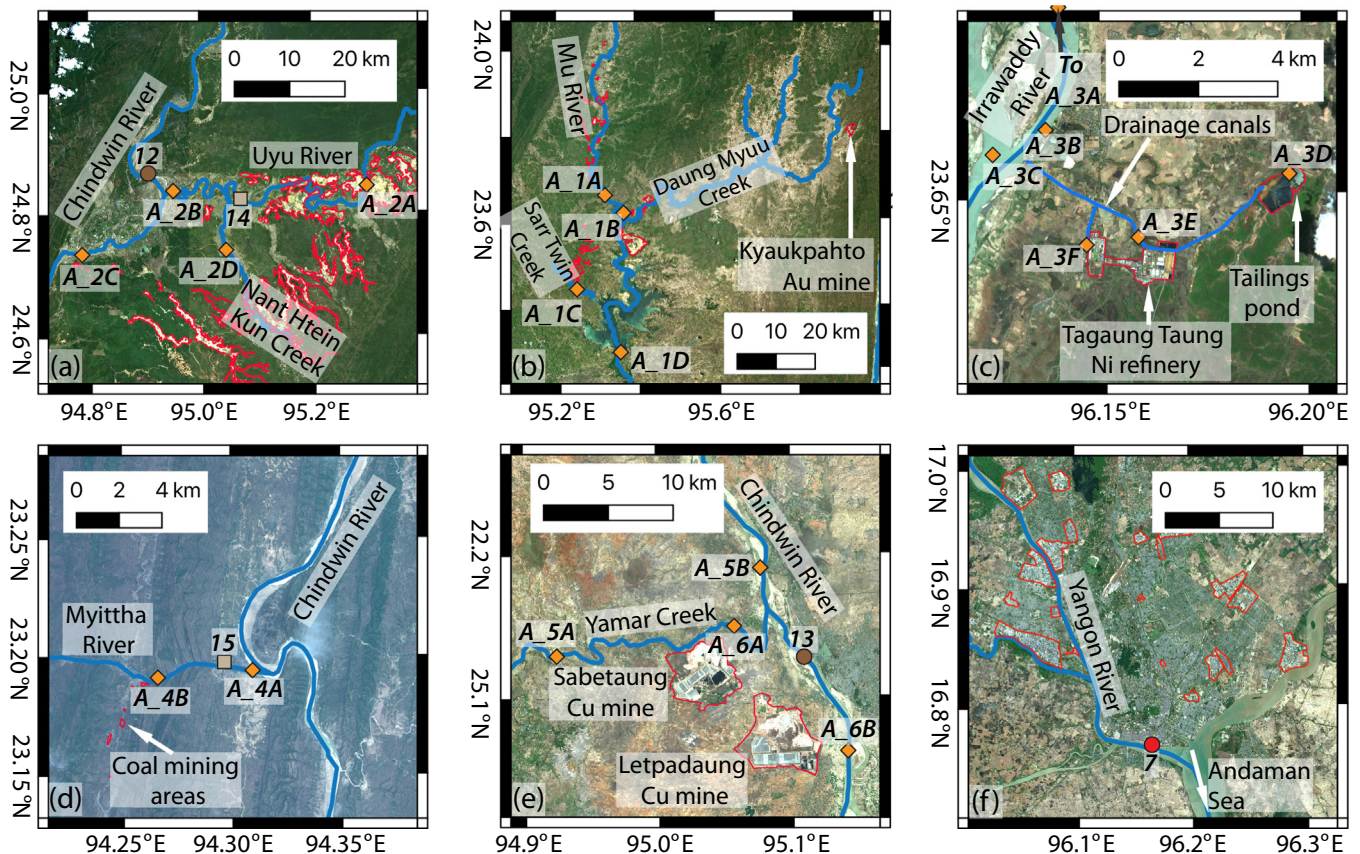


Fig. 2. Map of sampling locations close to major mining and industrial activities in the Irrawaddy River catchments. Locations of each of the panels in the Irrawaddy River catchment are shown as red boxes with letters in Fig. 1. Red outlines show areas of land disturbance by artisanal Au mining (panels (a) and (b)), Ni refinery facilities at Tagaung Taung (panel (c)), coal mining areas (panel (d)), large Cu mines (panel (e)) and industrial zones within the metropolitan area of Yangon (panel (f)). Maps show true color satellite images captured by the Copernicus Sentinel 2 mission (Main-Knorn et al., 2017) between the 28th April and 29th May, 2019.

2.1.2. Salween River catchment

The Salween River catchment covers 283,500 km², straddling China (48 %), Thailand (7 %) and Myanmar (44 %), with a population of over 10 million people (Johnston et al., 2017). It rises in Tibet, extending ~2800 km south draining into the Gulf of Martaban near the town of Hpa-An (Fig. 1). Annual average discharge for the Salween River is poorly constrained, but estimated to be ~149 km³ yr⁻¹, ranking it 26th globally (Baronas et al., 2020; Milliman and Farnsworth, 2011). Discharge follows a similar monsoon driven seasonal changes as the Irrawaddy River (Baronas et al., 2020). Unlike the Irrawaddy, the Salween River catchment features almost no flood plain/flat topography and is consequently much less densely populated (Johnston et al., 2017). The most densely populated region of the catchment is close to the mouth of the Salween River (Johnston et al., 2017). There are a variety of known metal ore deposits in the lower Salween River catchment, notably Sb, Sn-W, Pb-Zn-Ag deposits associated with the Mogok metamorphic belt and sedimentary rocks of the Shan plateau (Fig. 1) (Gardiner et al., 2014; Khin Zaw, 2017). Salween River water time-series samples were collected at a single station (station 16), close to the mouth of the river at the town of Hpa-An (Fig. 1, Supplementary Table 1).

2.1.3. Sittaung River catchment

The Sittaung River catchment (area; 35,000 km², length; 560 km, Milliman and Farnsworth, 2011) resides entirely within Myanmar (Fig. 1). With an annual average water discharge of ~35 to 45 km³ yr⁻¹, it is the fourth largest river in the country in terms of water discharge after the Irrawaddy, Chindwin and Salween rivers (Yamashita and Htay Aung, 2016). It has a relatively high population density, home to 5.8

million people, mostly residing along the central valley of the river catchment (Salmivaara et al., 2013; Nesheim et al., 2017; Johnston et al., 2017). There are a number of metal ore deposits within the catchment, predominantly Au deposits associated with the Mogok metamorphic belt (Fig. 1) (Gardiner et al., 2014; Ye Myint Swe et al., 2017). Time-series water samples from the Sittaung River were collected within the city of Naypyitaw (~924,000 people), the capital city of Myanmar, in the upper reaches of the catchment (station 17) (Supplementary Table 1).

2.1.4. Kaladan River catchment

The Kaladan River catchment covers 40,000 km² straddling India and Myanmar with a river length of 320 km (Milliman and Farnsworth, 2011) (Fig. 1). This river catchment has been subject to very limited previous study, with water discharge remaining poorly constrained. The Kaladan River catchment is largely unperturbed by anthropogenic activities, being the 5th largest catchment in the world that remains completely free from dam construction (Grill et al., 2019). The catchment has, however, been subject to major infrastructure development in recent years (Pandit and Basu, 2014). Time-series water samples were collected at Kyauktaw (station 18), close to the mouth of this river (Fig. 1, Supplementary Table 1).

2.2. Methods

2.2.1. Sample collection and analytical measurement

Water samples were collected using a plastic bucket from the center of the river channel accessed either by boat or from a bridge. Prior to collection, the plastic bucket was rinsed 3 times with the river water. Water temperature was measured using a Hanna Instruments (model

pHep4) temperature and pH meter. Between 30 and 60 mL of water was filtered immediately after collection through a 0.22 µm polyethersulfone capsule filter (Millex-GP, 33 mm diameter) using a syringe. Filtered water was collected into acid cleaned high-density polyethylene bottles. Prior to filtration the syringe was rinsed 3 times with river water, following which 10 to 15 mL of river water was flushed through the capsule filter to clean any potential contamination. Samples collected at stations 1, 3, 4, 5 and 13 were acidified to pH ~2 by the addition of ~30 µL of 16 M distilled HNO₃ in the field. Following collection, the sample bottles were stored in zip-lock plastic bags in the dark prior to shipment to the Department of Earth Sciences, University of Oxford (United Kingdom), typically within 3 months of sample collection. Within a clean room laboratory at the University of Oxford appropriate volumes of distilled 16 M HNO₃ were added to the samples to achieve a 2 % v/v HNO₃ solution. Blanks of the collection process were assessed by filtering and acidifying milli-q water following the same protocols outlined above.

Acidified samples were analyzed for trace element concentrations using a PerkinElmer NexION 350D quadrupole inductively coupled plasma mass spectrometer (ICP-MS) at the Department of Earth Sciences, University of Oxford. The instrument was calibrated from a series of standards that were robotically prepared from two stock solutions by a coupled Elemental Scientific prepFAST M5 autosampler. The stock solutions were prepared by mixing and diluting aliquots from a collection of synthetic ICP elemental standards (Merck Certipur - single element and custom blend) into a 2 % v/v HNO₃ solution on the day of analysis.

The ICP-MS was setup to measure all of the elements together in a single method using the PerkinElmer Syngistix ICP-MS software. This method also incorporated the use of the instrument's dynamic reaction/collision cell: technology that suppresses molecular interferences for large swathes of measured mass spectra, and is very effective in improving the analyte signal to background ratio for many elements. Supplementary Table 2 details the suite of elements that were analyzed and the cell mode that was adopted.

Once the instrument had been calibrated the samples were measured in groups of ten, which were bracketed with instrument blanks and a certified or in-house reference standard (either NRC Canada SLRS-6 River Water, or Irrawaddy River water sample from station 3, 27/08/2017) to check for memory effects and as a quality and consistency check, respectively. The long-term reproducibility of these reference standards, and the agreement of results for SLRS-6 to values certified by NRC Canada (Yang et al., 2015), or consensus values from Yeghicheyan et al., 2019 are detailed in the Supplementary Information file (Supplementary Table 3). Within each sample group every tenth sample was measured in duplicate to determine analytical precision. After thirty samples were measured the instrument was re-calibrated to minimize the possibility of analytical drift, caused by matrix effects and 'matrix build-up' on the sample introduction system as the run progressed. Most samples were measured without dilution. Those samples that did require dilution were diluted no more than ten times. Internal standards (In, Rh, Ir and Re) were doped into every blank, standard and sample by the prepFAST to normalize for any plasma suppression (Supplementary Table 2).

2.2.2. Calculation of discharge-elemental concentration relationships, elemental fluxes and yields

Sampling stations 1, 4, 5, 12, 13 and 16 are situated close to hydrological monitoring stations run by the Department of Meteorology and Hydrology (DMH), Myanmar (Fig. 1). Daily average river water discharge data collected at these hydrological stations was provided by DMH and were used to assess relationships between trace element concentrations and discharge (termed C-Q relationships, herein), following a power law (e.g. Baronas et al., 2017) (Eq. (1)).

$$C = aQ^b \quad (1)$$

where C denotes elemental concentration (µg/L), Q denotes daily average discharge (m³/second), and a and b are constants. Best fits to the exponent, b, and their 2σ uncertainties, along with adjusted-r² values and p values

for these relationships were obtained using the MATLAB curve fitting application.

Annual dissolved trace element fluxes were calculated for stations 5, 13 and 16. These stations capture the integrated fluxes from the Irrawaddy (station 5), Chindwin (station 13) and Salween (station 16) river catchments (Fig. 1). Monthly averaged trace element fluxes were calculated by multiplying mean trace element concentrations of samples collected in each calendar month by monthly averaged water discharge data for the years 2017, 2018 and 2019. Annual trace element fluxes were calculated by summation of these monthly averaged trace element fluxes. For these stations, 3 to 5 samples are available for the majority of the calendar months, suggesting a reasonable assessment of monthly variation. Notable exceptions are the months May and July for station 16 (Salween River) for which no samples are available. Monthly averaged trace element concentrations for these months were interpolated using values from the adjacent months. For values that were below the limit of quantification, the mid-range value of the limits of quantification encountered during the measurement sessions was used (Supplementary Table 3).

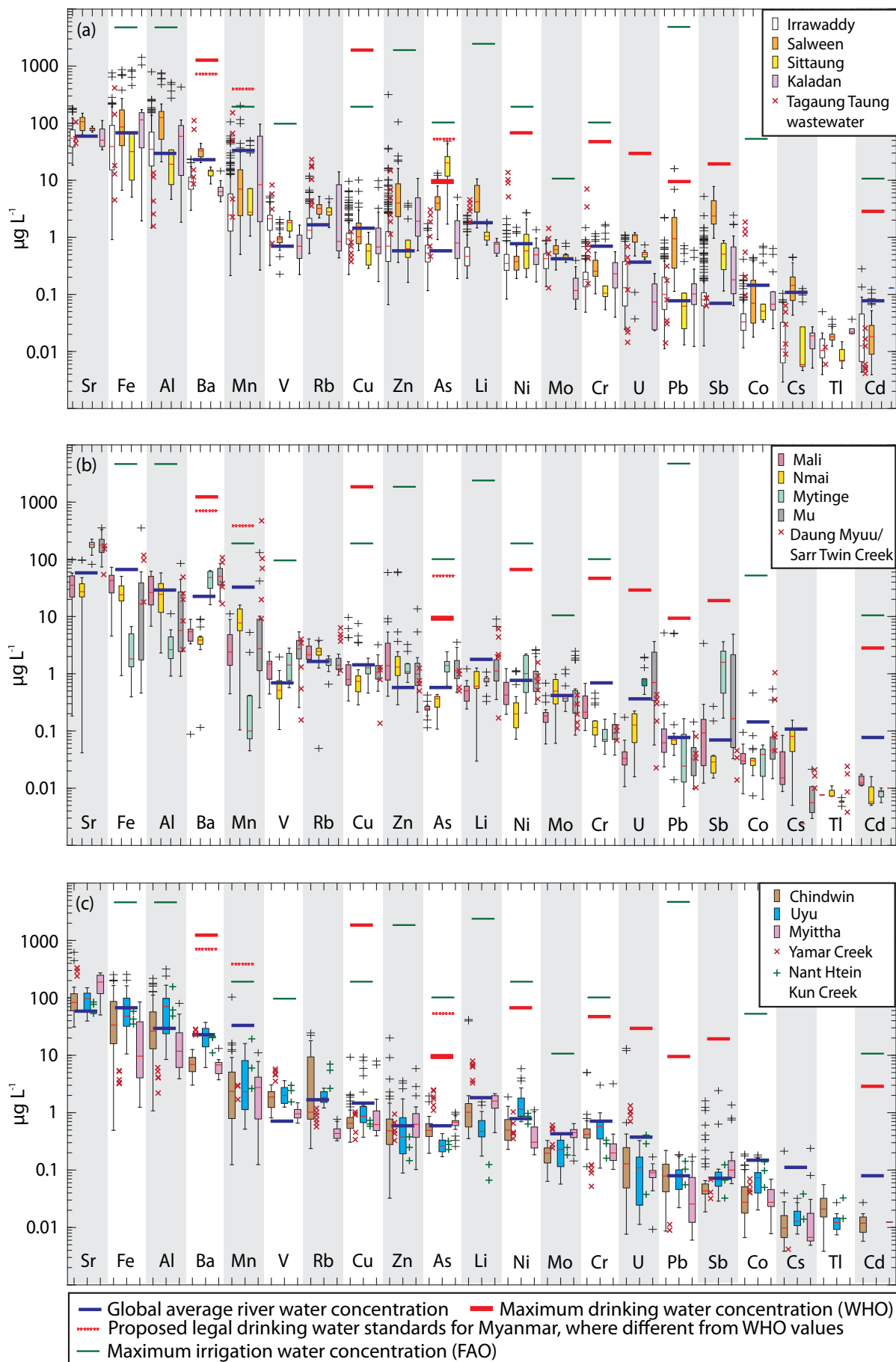
Annual yields for each element (annual fluxes normalized to catchment area; kg/year/km²) were calculated for the Irrawaddy, Chindwin and Salween river catchments. This assumed that the fluxes derived from data collected at stations 1, 13 and 16 represent total dissolved elemental fluxes exported from these catchments, with areas of 413,710 km² (Irrawaddy), 115,300 km² (Chindwin) and 283,500 km² (Salween) respectively (Ketelsen et al., 2017; Johnston et al., 2017; Shrestha et al., 2020).

Uncertainties on monthly average elemental fluxes (1σ) were calculated by propagation of the standard errors on monthly averaged elemental concentrations and discharges using Monte Carlo simulations. For months with <3 data points above the limits of quantification, uncertainties on the monthly averaged elemental concentrations were taken as the mean standard deviation of the months with >3 data points, as outlined in Chapman et al., 2015. Uncertainties on annual elemental fluxes (1σ) were calculated by propagation of uncertainties on mean monthly fluxes (1σ) by Monte Carlo simulation.

3. Results

Trace element concentration data produced in the study are accessible in the Electronic Data file and the Hydroshare data repository (Bridgestock et al., 2022). Interquartile ranges of elemental concentrations in the studied catchments largely agree with, or are lower than, average values from a global compilation of large rivers catchments thought to be unperturbed by anthropogenic activities (Fig. 3) (Gaillardet et al., 2014). Notable exceptions include As in the Sittaung River, as well as As, Pb, Zn and Sb in the Salween River, which are all significantly higher than global average river water concentrations. The Mu and Myitnge rivers are also enriched in Sb relative to global average river water concentrations. Trace element concentrations are predominantly below maximum values for drinking water standards proposed for Myanmar (Oo and Hata, 2019), and those recommended by the World Health Organization (WHO) (WHO, 2017). They are also lower than water standards for irrigation water recommended by the Food and Agriculture Organization (FAO) of the United Nations (Svobodová et al., 1993). Exceptions are As in the Sittaung River (station 17), with 14 out of 18 samples featuring As concentrations higher than the safe drinking water limit recommended by the WHO of 10 µg/L (WHO, 2017) (Fig. 4). In the Salween River (station 16), 4 and 2 out of 33 samples are also higher than the safe drinking water limits recommended by the WHO for As and Pb respectively (10 µg/L for both elements) (WHO, 2017). The Mn concentration of 1 out of the 2 samples collected from Sarr Twin Creek is also higher than the proposed drinking water standard for Myanmar, and FAO standards for irrigation water.

Trace element concentrations in samples collected close to mining and industrial activities (Fig. 2), for the most part, are not significantly enriched in trace element concentrations relative to either global average values (Gaillardet et al., 2014) or samples collected in the Irrawaddy River catchment at locations distal to such human activities (Fig. 3, Supplementary



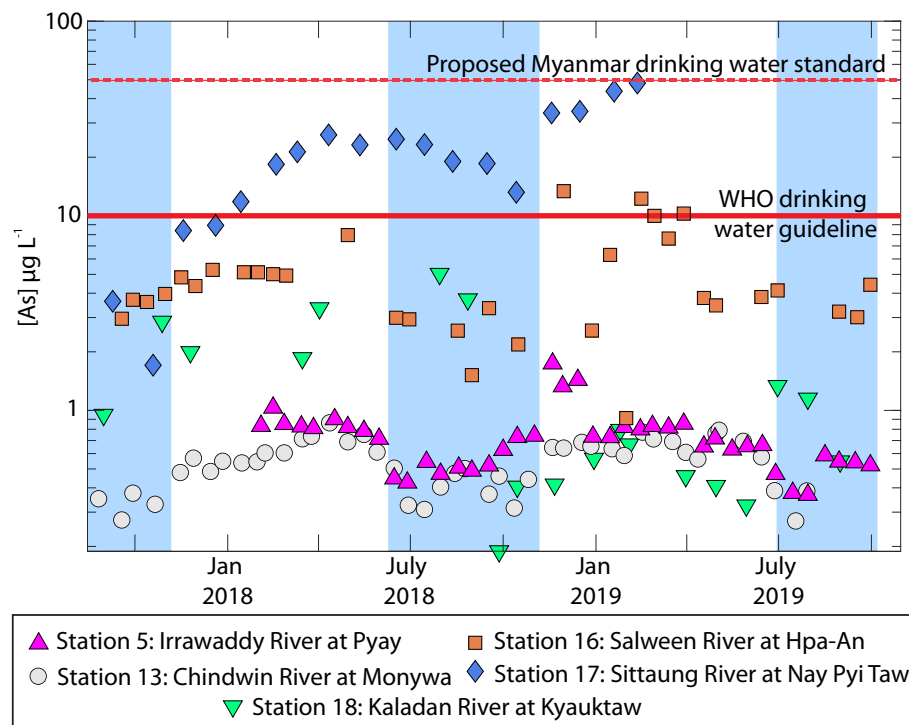


Fig. 4. Comparison of time-series of dissolved river water As concentrations for the 5 major river catchments sampled in this study. Solid and dashed red lines denote the WHO drinking water guideline and proposed Myanmar drinking water standards for As concentrations (WHO, 2017, Oo and Hata, 2019). Blue shaded regions denote periods of high river water discharge associated with the monsoon season.

Figs. 3 and 5 to 8). Exceptions include wastewater from the Tagaung Taung Ni refinery, which is enriched in Ni, As, Li, Cr, Co and Rb compared to the Irrawaddy River (Fig. 3, Supplementary Fig. 3). River water collected in Yamar Creek also shows a factor of ~ 2 increase in Cu concentrations between locations upstream and downstream of the Sabetaung Cu mine (Fig. 2e, Supplementary Fig. 8). Despite this increase, Cu concentrations in this river (up to $0.99 \mu\text{g/L}$) remain lower than the global average for river water (of $1.48 \mu\text{g/L}$; Gaillardet et al., 2014).

Concentrations of individual elements vary by a factor of ~ 2 to ~ 1000 at a given station (Fig. 5, Supplementary Figs. 2 to 9). For many elements this variance is linked to changes in water discharge (Eq. (1), Fig. 6). The exact behavior of individual elements varies between stations, but elements can be broadly divided into 3 groups based on their C-Q relationships (Eq. (1)). The elements Fe, Al, Ni, Co, Cr and Pb typically increase concentration with discharge, characterized by $b > 0$ in Eq. (1) (Fig. 6), with higher concentrations during the monsoon season (Fig. 5, Supplementary Figs. 2 to 9). Conversely, Ba, As, Mo, Sr, Li, U, Sb and Cd tend to decrease with discharge, characterized by $b < 0$ Eq. (1), with higher concentrations during the dry season. The elements Cu, Zn, Cs, Rb, Tl, V and Mn tend to display either constant concentrations ($b = 0$, Eq. (1)) or variance that is not related to changing discharge.

Annual elemental fluxes and yields for the Irrawaddy (station 5), Salween (station 16) and Chindwin River (station 13) catchments are

given in Table 1, with monthly averages given in the Electronic Data file, and shown in Supplementary Fig. 10. Annual elemental fluxes by the combined dissolved loads of the Irrawaddy and Salween rivers, sampled close to their mouths (stations 5 and 16) represent between $\sim 1\%$ and 5% of global estimates for most elements (Gaillardet et al., 2014). Zinc, Pb and Sb fluxes are still more significant and their combined annual dissolved fluxes from the Irrawaddy and Salween rivers represent between $\sim 8\%$ and 17% of global estimates. These high fluxes are dominated by fluxes from the Salween River that features particularly high concentrations of these elements.

Yields (area normalized fluxes) of most elements for the Irrawaddy, Chindwin and Salween river catchments are between 2 and 10 times higher than global averages, while yields of Zn, Pb and Sb for the Salween River are 16, 31 and 60 higher than global averages respectively (Fig. 7). Manganese and Cd are also exceptions to these observations featuring yields significantly lower than global averages.

4. Discussion

4.1. Global importance of dissolved trace element fluxes transported by the Irrawaddy and Salween rivers

The Irrawaddy and Salween represent 2 of the 10 major rivers catchments draining the Himalayan mountain belt. The combination of high

Fig. 3. Summary of trace element river water concentration results for different river catchments. Boxes represent the interquartile range, whiskers the full range (excluding outliers) and the thin red bars the median of results for each catchment. Outliers are shown as black crosses, defined as values that deviate outside the interquartile range by more than 1.5 times the interquartile range. Data for Yamar Creek, Nant Htein Kun Creek, Daung Myuu Creek, Sarr Twin Creek and wastewater draining/tailings pond from Tagaung Taung (Fig. 2) are plotted as discrete data points due small sample populations ($n < 7$). Global average trace element concentrations are from Gaillardet et al. (2014). WHO and proposed Myanmar drinking water standards are from WHO (2017) and (Oo and Hata, 2019) respectively. Irrigation water standards water recommended by the Food and Agriculture Organization of the United Nations (FAO) are from Svobodová et al. (1993). A significant number of samples were below the limits of quantification for Cs, Tl and Cd, likely skewing the plotted box and whiskers to higher values. In panel (a), boxes for the rivers Salween, Sittaung and Kaladan are defined by data from stations 16, 17 and 18 respectively. Data from stations 1 to 7, and A_3A, A_3B and A_3C define the box for the Irrawaddy. In panel (b), boxes for the rivers Mali, Nmai, Mytinge and Mu are defined by data from stations 8, 9, 10 and 11, A_1A and A_1D respectively. In panel (c), data from stations 12, 13, A_2C, A_5B and A_6B define Chindwin boxes, stations 14, A_2A and A_2B define Uyu boxes and stations 15, A_4A and A_4B for Myiththa boxes.

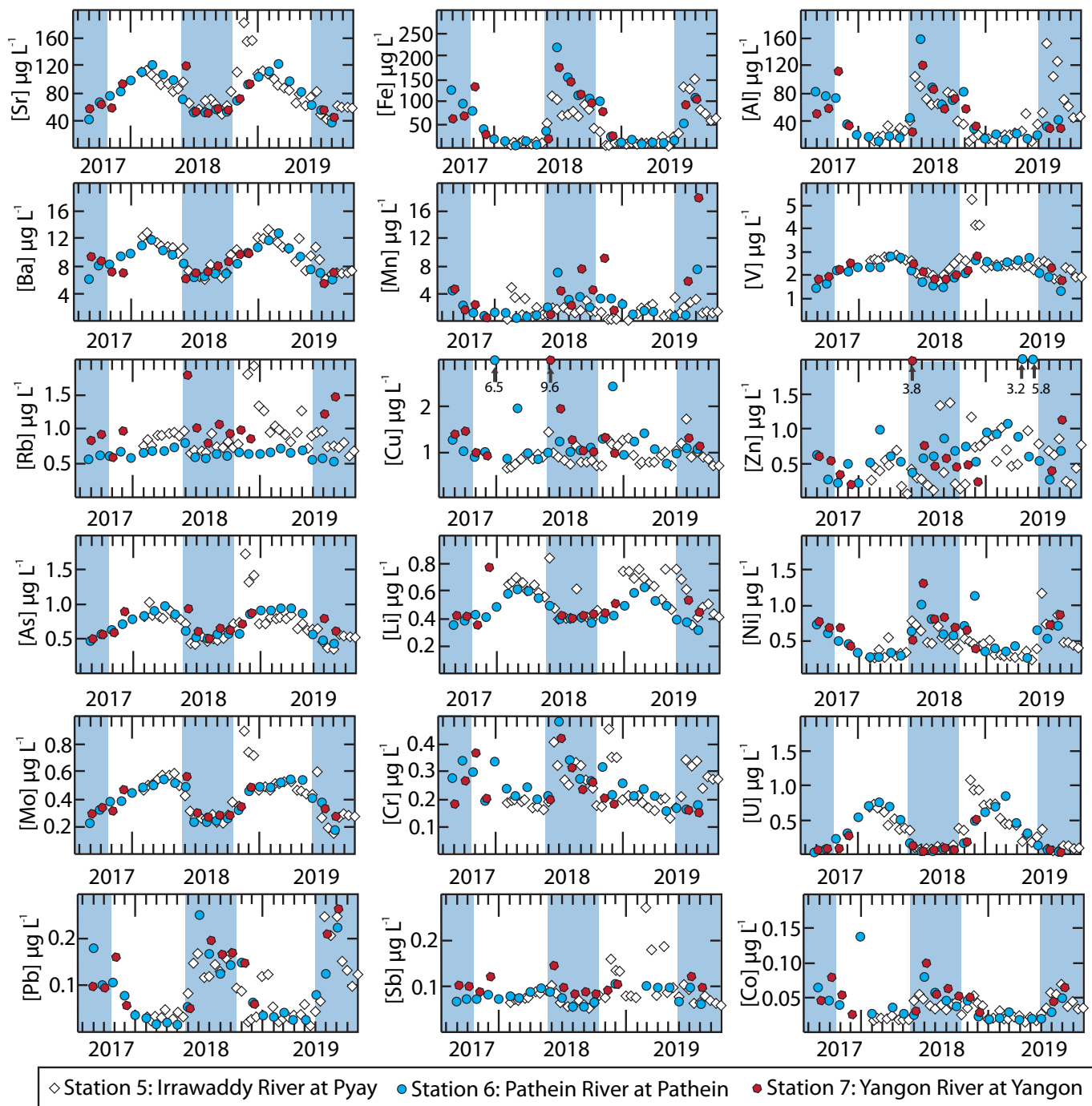


Fig. 5. Time-series trace element results for sampling stations within the lower Irrawaddy River catchment. Blue shaded regions show periods of high river discharge based on discharge data for the Irrawaddy River at Pyay from the Department of Meteorology and Hydrology, Myanmar. Note Tl and Cd were below the limits of quantification in all samples from these stations. Time-series results for all other stations are shown in Supplementary Figs. 2 to 9. Due to the influence of mixing with seawater during low discharge periods, trace element concentrations are only available during the high-discharge periods for station 7.

rates of tectonic uplift and warm, wet climatic conditions in this region result in high rates of both chemical and physical denudation (Chapman et al., 2015; Baronas et al., 2020). Consequently, the Irrawaddy and Salween play a key role in global biogeochemical cycles, ranking within the top ten rivers globally for fluxes of both solid and dissolved material to the ocean (Robinson et al., 2007; Bird et al., 2008; Furuichi et al., 2009; Milliman and Farnsworth, 2011; Chapman et al., 2015; Baronas et al., 2020). Our results further highlight the important role of these catchments in global biogeochemical cycles for their significant dissolved riverine trace element fluxes (Table 1). The combined annual dissolved fluxes from the Irrawaddy and Salween river catchments represent ~1 to 5 % of

global riverine flux estimates to the land-ocean interface for the majority of elements (Gaillardet et al., 1999). The dissolved fluxes of Zn, Pb and Sb of these rivers are yet more significant, contributing between 8 and 17 % of global flux estimates. We acknowledge that net dissolved elemental fluxes from these rivers to the ocean are likely to be modified by estuarine processes (e.g. Mosley and Liss, 2020).

The magnitude of these flux estimates (~1 to 5 % of global values) comes despite these catchments only covering 0.66 % of the global exorheic land area (Syvitski et al., 2005; Ketelsen et al., 2017; Johnston et al., 2017). Area normalized fluxes (yields) for most elements, are therefore ~2 to 10 times higher than global averages (Fig. 7). Large uncertainty in global

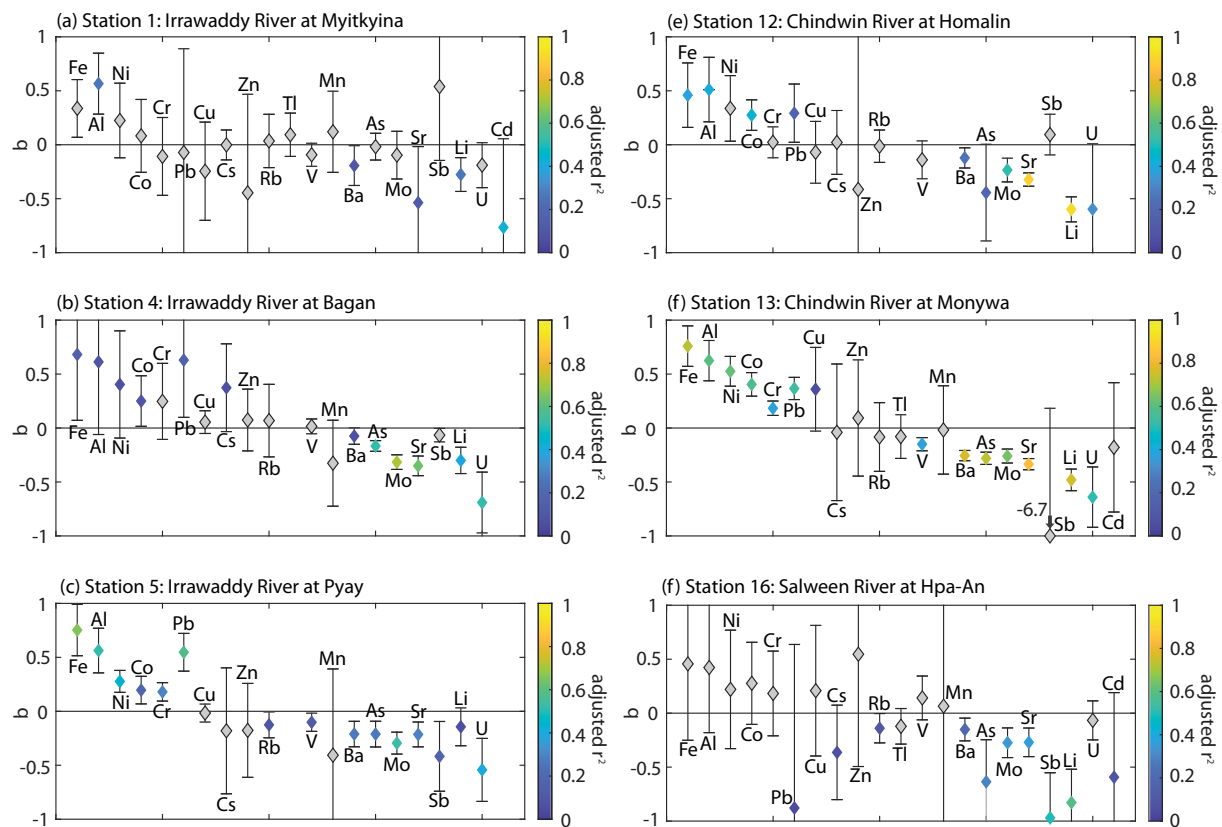


Fig. 6. Summary of elemental-concentration discharge relationships for stations 1, 4, 5, 12, 13 and 16 (Eq. (1)). Exponents of the power law relationships ('b') between discharge and elemental concentrations are plotted on the y-axis. The color scale denotes the adjusted- r^2 values of the relationships. Grey symbols show relationships with a p value > 0.05.

riverine trace element flux estimates requires such comparisons to be treated with caution. These observations are, however, consistent with chemical weathering rates being ~3 to 4 times higher than global averages in these catchments, as revealed by fluxes of major dissolved solutes from these rivers (Chapman et al., 2015). Yields for the Irrawaddy, Salween and Chindwin River catchments also are comparable to those for the Ganga River catchment (Borai et al., 2020), another large river catchment draining the Himalayan mountain range with a similar chemical weathering regime in terms of climate and tectonics (Table 1; Fig. 7).

Seasonal flux changes for most elements scale with discharge, with highest values during the monsoon season (Supplementary Fig. 10). The magnitude of these seasonal changes however varies for different elements, depending on their C-Q relationships (Eq. (1); Fig. 6). For elements with positive C-Q relationships (e.g. Fe, Al, Ni, Co, Cr and Pb at station 5, Pyay), fluxes increase by a factor ~20 to 140 during the monsoon season (June to October), with these months accounting for 82 to 96 % of the annual elemental fluxes (Supplementary Fig. 10). By contrast, for elements with negative C-Q relationships (e.g. Sr, Ba, V, As, Mo), fluxes only increase by a factor ~5 to 15 during the monsoon season, or in extreme cases (e.g. U, Sb, Li; $b \approx -1$), remain relatively constant throughout the year.

Differences in C-Q relationships are broadly interpreted in terms of changes in water flow pathways within the critical zone (e.g. Godsey et al., 2009; Torres et al., 2015). Negative C-Q relationships reflect the importance of solute sourcing from waters circulating at depth within the critical zone that get diluted by shallower runoff, behavior which is typical for major dissolved cations (e.g. Godsey et al., 2009). Trace elements displaying positive C-Q relationships have been linked to their association with colloidal phases, which get mobilized from surface layers within soils during periods high discharge (Trostle et al., 2016). We do not have data from ultra-filtration techniques to confirm colloidal associations for the trace elements displaying positive C-Q relationships in this study, but

these elements (Fe, Al, Ni, Co, Cr and Pb) are commonly observed to be associated into colloidal phases in other rivers (Pokrovsky et al., 2010; Nystrand et al., 2012; Trostle et al., 2016; Cuss et al., 2018).

4.2. Limited anthropogenic perturbation of trace elements in major Myanmar rivers

Several observations suggest that the dissolved trace element budgets are, in general, not significantly perturbed by anthropogenic inputs in the studied rivers. Firstly, the median concentrations of the majority of trace elements in the studied rivers are comparable to, or lower than, average values for other large rivers thought to be largely unperturbed by anthropogenic activities (Gaillardet et al., 2014, Fig. 3). Secondly, the ~2 to 10 times higher annual yields for the majority of elements from the Irrawaddy, Chindwin and Salween river catchments relative to global averages (Fig. 7, Table 1; Gaillardet et al., 2014) can be feasibly explained by the naturally higher rates of chemical weathering in these catchments constrained by major cation fluxes (Chapman et al., 2015). Thirdly, samples close to major mining/industrial activities in the Irrawaddy River catchment (Fig. 2), for the most part, are not significantly enriched in trace element concentrations compared to either global average river values (Gaillardet et al., 2014), or samples collected at locations in these catchments distal to such human activities (Figs. 3, 5, Supplementary Figs. 2 to 9). We elaborate on this final point below. Notable exceptions to these observations are As in the Sittaung River, and Sb, Pb and Zn in the Salween River (Figs. 3, 4 and 7). The potential anthropogenic sources of these elements in these rivers and water quality implications are discussed in Section 4.3.

The most densely populated region of the Irrawaddy River catchment is the Irrawaddy delta (Salmivaara et al., 2013); host to Yangon (5 million people) and Patheingyi (235,000 people) the 1st and 6th largest cities

Table 1

Annual dissolved trace element fluxes and yields from the Irrawaddy, Salween and Chindwin river catchments compared to global flux estimates (Gaillardet et al., 2014) and to those from the Ganges River catchment (Borai et al., 2020).

Element	Global ^a		Irrawaddy River catchment		Chindwin River catchment		Salween River catchment		Ganges River catchment ^b	
	Flux (kt/yr)	Yield (kg/yr/km ²)	Flux (kt/yr)	Yield (kg/yr/km ²)	Flux (kt/yr)	Yield (kg/yr/km ²)	Flux (kt/yr)	Yield (kg/yr/km ²)	Flux (kt/yr)	Yield (kg/yr/km ²)
Sr	2240	21	30 ± 1	72 ± 2	10.0 ± 0.5	86 ± 5	15.9 ± 0.6	56 ± 2	90 ± 41	92 ± 41
Fe	2470	23	30 ± 2	72 ± 5	16 ± 2	138 ± 15	34 ± 10	123 ± 36		
Al	1200	11	26 ± 2	63 ± 4	13 ± 1	113 ± 13	35 ± 9	125 ± 33	6.5 ± 5	6.6 ± 5
Ba	860	8	3.6 ± 0.1	8.7 ± 0.3	0.86 ± 0.05	7.5 ± 0.4	5.1 ± 0.2	18 ± 1	30 ± 13	31 ± 13
Mn	1270	12	0.83 ± 0.06	2 ± 0.2	0.45 ± 0.07	3.9 ± 0.6	3.5 ± 1.6	12 ± 6	0.15 ± 0.05	0.15 ± 0.05
V	27	0.25	0.98 ± 0.03	2.36 ± 0.04	0.27 ± 0.02	2.3 ± 0.2	0.18 ± 0.01	0.63 ± 0.05	2.6 ± 0.6	2.7 ± 0.7
Rb	61	0.58	0.36 ± 0.02	0.87 ± 0.04	1.0 ± 0.2	8.6 ± 1.6	0.52 ± 0.03	1.8 ± 0.1	2.2 ± 0.9	2.7 ± 0.7
Cu	55	0.52	0.38 ± 0.02	0.93 ± 0.04	0.18 ± 0.06	1.6 ± 0.5	0.29 ± 0.08	1.0 ± 0.3	1.1 ± 0.3	1.2 ± 0.4
Zn	23	0.22	0.26 ± 0.05	0.64 ± 0.13	0.11 ± 0.03	1.0 ± 0.2	1.6 ± 0.8	6 ± 3		
As	23	0.22	0.26 ± 0.01	0.62 ± 0.03	0.061 ± 0.004	0.52 ± 0.03	0.69 ± 0.08	2.4 ± 0.3	2.5 ± 1.0	2.5 ± 1.0
Li	69	0.65	0.23 ± 0.01	0.56 ± 0.03	0.10 ± 0.01	0.87 ± 0.06	0.64 ± 0.05	2.3 ± 0.2	2.5 ± 1.7	2.6 ± 1.7
Ni	30	0.28	0.22 ± 0.01	0.53 ± 0.03	0.11 ± 0.01	0.95 ± 0.08	0.09 ± 0.02	0.31 ± 0.05	1.1 ± 0.3	1.1 ± 0.3
Mo	16	0.15	0.14 ± 0.01	0.33 ± 0.03	0.025 ± 0.002	0.21 ± 0.02	0.092 ± 0.04	0.33 ± 0.01		
Cr	26	0.25	0.114 ± 0.004	0.28 ± 0.01	0.074 ± 0.006	0.64 ± 0.05	0.07 ± 0.01	0.24 ± 0.04	0.3 ± 0.2	0.3 ± 0.2
U	14	0.13	0.092 ± 0.008	0.22 ± 0.02	0.013 ± 0.001	0.12 ± 0.01	0.16 ± 0.01	0.55 ± 0.04		
Pb	3	0.028	0.054 ± 0.004	0.13 ± 0.01	0.020 ± 0.002	0.17 ± 0.01	0.25 ± 0.13	0.9 ± 0.5	0.25 ± 0.05	0.25 ± 0.05
Sb	2.6	0.025	0.032 ± 0.003	0.08 ± 0.01	0.02 ± 0.02	0.20 ± 0.20	0.41 ± 0.05	1.5 ± 0.2		
Co	5.5	0.052	0.017 ± 0.001	0.04 ± 0.002	0.0080 ± 0.001	0.070 ± 0.007	0.02 ± 0.01	0.09 ± 0.02	0.1 ± 0.05	0.1 ± 0.05
Cs	0.4	0.0038	0.004 ± 0.002	0.009 ± 0.006	0.0019 ± 0.0002	0.017 ± 0.012	0.025 ± 0.04	0.09 ± 0.01		
Tl	NA	NA					0.0031 ± 0.0002	0.011 ± 0.001		
Cd	3	0.028					0.004 ± 0.001	0.013 ± 0.003	0.05 ± 0.005	0.05 ± 0.005

^a Global flux values are taken from Gaillardet et al. (2014), with global yields calculated by normalizing these values to a global exorheic land area of 106,000,000 km² (Syvitski et al., 2005).

^b Fluxes and yields for the Ganges River catchment are calculated using mean (± 1sd) dissolved trace element concentrations from time-series data at Manikchak published by Borai et al. (2020), and an annual discharge and catchment area of 490 km³/year and 980,000 km² respectively (Milliman and Farnsworth, 2011).

in Myanmar respectively. Comparison of time-series data for stations 5, 6 and 7 provide insights on the impact of trace element pollution from municipal and industrial waste discharge to rivers in this densely populated region (Fig. 5). Station 5 samples the main Irrawaddy River channel upstream of the Irrawaddy delta, while stations 6 and 7 sample two of the delta distributaries at Pathein and Yangon respectively (Fig. 1). Notably, station 7 is situated on the Yangon River downstream of most of the municipal development in Yangon and several industrial zones (Fig. 2f). Time-series results from stations 5, 6 and 7 are in good agreement for all measured dissolved trace element concentrations (Fig. 5). This suggests that the dissolved riverine budgets studied trace elements are not significantly perturbed by inputs from municipal and industrial sources in these densely populated regions of the catchment. Note that data is only available during periods of river water high discharge (June to December) at station 7 to make such a comparison. During periods of low discharge (January to May) water chemistry at this location is impacted by mixing with seawater.

Samples of rivers draining areas heavily impacted by mining are generally not enriched in trace elements relative to locations distal to such activities (Fig. 3, Supplementary Figs. 2 to 9). Wastewater from the Taguang Taung Ni-Cr ore refinery facility is enriched in several elements (Ni, As, Li, Cr, Co and Rb). Samples from the adjacent section of the Irrawaddy River however show that this pollution does not significantly impact Irrawaddy River dissolved elemental fluxes (Fig. 2c, Supplementary Fig. 3). Likewise sampling along a transect of Yamar Creek indicates Cu inputs from the nearby Sabetaung Cu mine, but that these inputs do not significantly impact dissolved Cu fluxes transported by the Chindwin River (Fig. 2e, Supplementary Fig. 8). Finally, samples close to both coal mining activities in the Myittha River catchment (Fig. 2d), and artisanal Au mining in the Uyu and Mu river catchments (Fig. 2a, b) show no clear sign of dissolved trace element enrichment relative to other locations in these river systems (Supplementary Figs. 5 to 7). Note that release of Hg to rivers, related to ore processing techniques used by artisanal Au mining operations (Esdaile and Chalker, 2018), is a concern for human health (e.g. Berzas Nevado et al., 2010). We are not able to assess such pollution and associated water quality issues within the context of this current study, given the specialist collection and analytical techniques required for dissolved Hg concentration measurements (Leopold et al., 2010).

In summary, with some notable exceptions (Section 4.3) dissolved riverine trace element fluxes in the studied catchments currently appear unperturbed by human activities. The presented data should therefore serve as a useful ‘natural’ baseline, against which future perturbations to trace element fluxes in these rivers, arising from either human developments (in particular of the mining industry) or climate change, could be assessed.

4.3. Possible anthropogenic perturbations to riverine trace element concentrations and water quality implications for human exposure

4.3.1. Arsenic in the Sittaung River

Dissolved As concentrations in the Sittaung River (station 17) are a water quality concern in this catchment. Fourteen out of 18 samples from this station have As concentrations higher than the safe drinking water limit recommended by the WHO, of 10 µg/L (Fig. 4; WHO, 2017). The highest two As concentrations (44 and 48 µg/L) approach the proposed Myanmar drinking water standard of 50 µg/L, and are about half that recommended for irrigation water (Svobodová et al., 1993).

Sittaung River As concentrations are anomalously high when compared to those typically observed in rivers of <1 µg/L (Fig. 4, Smedley and Kinniburgh, 2002, Gaillardet et al., 2014). The cause of the high As concentrations observed in the Sittaung River is unclear. Based on studies of other As enriched rivers, potential candidates include natural inputs from geothermal springs or As enriched groundwater, or anthropogenic inputs from municipal waste (sewage) or discharge from mine tailings (Smedley and Kinniburgh, 2002, Wang and Mulligan, 2006, Cheng et al., 2009).

There are geothermal springs in the upper reaches of this catchment associated with Sb mineralization (Toe Aung Kyaw, 2017), which could be a possible source of As (and Sb; Fig. 3) enrichment in this river. Groundwater sampled in aquifers in the lower reaches of this catchment have been shown to have high As concentrations of ~14 to 36 µg/L (Pincetti-Zúñiga et al., 2020), similar to the observed Sittaung River water concentrations (~1.7 to 48.1 µg/L). However, inputs from both of these natural sources would be expected to be diluted by high river water discharge during the monsoon season, which does not fit with the observed temporal As concentration variations (Fig. 4). The Sittaung River sampling station is located within the city of Nay Pyi Taw (population ~924,000 people). Discharge

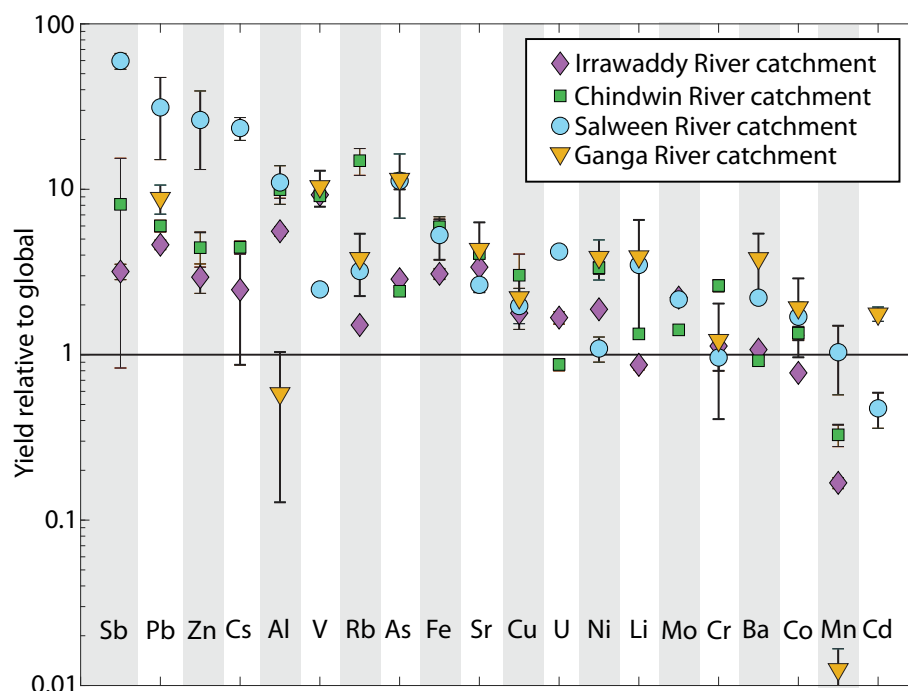


Fig. 7. Annual elemental yields (area normalized fluxes, kg/km²/year) for the Irrawaddy, Chindwin, Salween and Ganga river catchments normalized to global estimates (Table 1; Gaillardet et al., 2014). Global average elemental yields were calculated by dividing the global riverine elemental flux estimates of Gaillardet et al. (2014) by a global exorheic land area of 106,000,000 km² (Syvitski et al., 2005). Values for the Ganga River catchment were calculated using data from Borai et al. (2020).

of municipal waste could conceivably be a key source of As to these river waters. There are also a number of mining operations in this catchment upstream of this sampling station (Fig. 1), which could be potential sources of As to this river. Notably, these include Modi Taung, a large Au deposit containing As enriched pyrite (Ye Myint Swe et al., 2017), and Sb deposits that are known to be enriched in As (Toe Aung Kyaw, 2017). Given the importance of the Sittaung River as a water source to the 5.8 million people in this catchment (Nesheim et al., 2017) investigating these potential As sources should be considered a priority for future assessment.

4.3.2. Antimony, As, Pb and Zn in the Salween River

The Salween River, sampled at the town of Hpa-An (station 16), is significantly enriched in dissolved Sb, As, Pb and Zn relative to global averages of rivers that are thought to be largely unperturbed by anthropogenic activities (Fig. 3; Gaillardet et al., 2014). Furthermore, yields of these elements are between 11 and 60 times higher than global averages (Fig. 7, Electronic Data), significantly higher than relative yields of major cations derived from rock weathering (Chapman et al., 2015). In terms of water quality, dissolved Sb concentrations remain ~4 times lower than recommended safe drinking water limits but 4 and 2 out of the 33 samples are above the safe drinking water limits recommended by the WHO for As and Pb respectively (Figs. 3, 4, WHO, 2017).

The exact sources and processes resulting in elevated dissolved concentrations and area normalized fluxes of Sb, As, Pb and Zn in the Salween River, relative to other large global rivers (Gaillardet et al., 2014) including the Irrawaddy (this study), remain uncertain. The Salween River catchment, however, is host to a numerous Sb, Pb-Zn-Ag, Cu and Au ore deposits (Fig. 1; Toe Aung Kyaw, 2017, Than Htun et al., 2017a, Khin Zaw et al., 2017, Ye Myint Swe et al., 2017). Furthermore, soils in the upper reaches of this catchment in Yunnan province (China) contain elevated Sb concentrations, attributed to abundant Sb mineralization in this region (He et al., 2012). Mobilization of these trace elements from ore deposits, either due to natural chemical weathering or as a result of mining activities, may therefore explain their elevated dissolved concentrations and fluxes in the Salween River.

5. Conclusions

A large time-series dataset for dissolved trace element concentrations are presented for globally important, but chronically understudied river catchments, leading to the following conclusions. The Irrawaddy and Salween rivers play a major role in global trace element biogeochemical cycles; supplying ~1 to 17 % of global riverine dissolved trace element fluxes to the land-ocean interface, with area normalized fluxes ~2 to 10 times higher than global averages for most elements. These higher fluxes can be explained by enhanced rates of chemical weathering in these catchments. With some exceptions, trace element fluxes in the Irrawaddy, Salween, Sittaung and Kaladan rivers remain largely unperturbed by anthropogenic activities. The presented dataset should therefore provide 'natural' baselines of trace element fluxes in these rivers, against which future perturbations due to climate change and/or the development of Myanmar's mining industry can be assessed. Arsenic in the Sittaung River and Sb, As, Pb and Zn in the Salween River are notable exceptions that may already be perturbed by mining activities in these catchments. Elevated concentrations of As in the Sittaung River represent a water quality concern with potential to impact human health. The source of this As contamination should be considered a priority for future environmental research in this region.

CRedit authorship contribution statement

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Gideon M. Henderson: Conceptualization, Supervision, Funding acquisition, Writing-Review & Editing.

Phil Holdship: Methodology, Investigation, Writing-Review & Editing. Aung Myo Khaing: Project Administration, Resources.

Tin Tin Naing: Project Administration.

Tin Aung Myint: Investigation, Writing-Review & Editing.

Wint Wint Htun: Investigation, Writing-Review & Editing.

Win Khant: Investigation.

Win Myo Thu: Conceptualization, Project Administration.

Mo Aung Nay Chi: Project Administration.
 J. Jotautas Baronas: Writing-Review & Editing, Methodology.
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 Hazel Chapman: Methodology.
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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.156756>.

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