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Identifying global trends and drivers of freshwater aluminium concentrations using GloFAD (Global Freshwater Acidification Database)

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Aluminum is toxic to most aquatic and terrestrial organisms. Increased Al concentrations in soils and freshwaters are a direct result of human activity, via increases in acid deposition. Elevated Al concentrations pose a wide variety of threats to ecosystems and society, from causing human neurotoxicity, reducing carbon sequestration in forests, threatening biodiversity, and increasing the cost of water treatment. Freshwater aluminium concentrations increased across Europe and North America between the 1960s and 1990s, predominantly due to ecosystem acidification. Following acidic deposition reduction legislation enacted in the 1990s, the problems of acidification and increased freshwater aluminium concentrations were considered solved. However, recently and unexpectedly, Sterling et al. identified aluminum concentrations to be increasing across North America and Scandinavia. Sterling et al. proposed a conceptual model suggesting these widespread increases in freshwater aluminium concentrations resulted from a hysteresis of base cation and dissolved organic carbon (DOC) response to decreasing acidic deposition, where base cation increase is slow compared to that of DOC, resulting in elevated freshwater aluminium concentrations. This process can be exacerbated by further increases in DOC due to increasing global surface temperatures. The Sterling et al. conceptual model is supported by prior work by Weyhenmeyer et al. (2019, Scientific Reports) and Monteith et al., 2007, Nature) who identified widespread decreasing calcium and increasing DOC concentrations. In this study, we aim to validate the Sterling et al. model and identify if it is generalizable to other regions with decreasing calcium and increasing DOC trends, irrespective acidification status. Additionally, we aim to characterize other regions across the globe which are at risk of elevated aluminium concentrations. To fulfill these research goals, we compiled a large-sample water chemistry database from existing national and global datasets – GloFAD (Global Freshwater Acidification Database). The database is comprised of over 11 million unique samples spanning nearly 286,000 sites located between Antarctica and Russia, 18 years (2000 to 2019), and 40 water chemistry parameters. Preliminary analysis shows that aluminium is increasing in 27% to 71% of sites, dependent on the species, base cations are decreasing for 62% to 70% of sites, freshwater organic carbon is increasing for 58% to 64% of sites, and water temperature is increasing in 61% of sites. Increasing dissolved aluminium trends are strongly significantly correlated with decreasing base cation trends (calcium $\tau = -0.71$ and magnesium $\tau = -0.59$, $\alpha < 0.05$) but not with DOC concentrations ($\tau = -0.08$). The lack of correlation with DOC indicates that drivers of increasing

aluminium trends may differ based on the acidification status of the watershed and that regional models of freshwater aluminium chemistry may not be globally applicable. The widespread decreasing base cation trends and strong correlation between decreasing base cation and increasing aluminium trends indicates that increasing aluminium concentrations may become more widespread, posing a threat to aquatic and terrestrial organisms, potentially including humans, reducing carbon sequestration in forests, threatening biodiversity, and increasing water treatment costs.