Earth's Future

RESEARCH ARTICLE
10.1029/2021EF002217

A Cross-Cutting Approach for Relating Anthropocene, Environmental Injustice and Sacrifice Zones

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Key Points:

- Geochemical data provide a science-based tool to inform the discussion on Anthropocene governance in severely degraded territories
- In our case study, socio-environmental inequalities are linked to economic and governance processes operating from local to global scales
- Policies embracing local environmental rehabilitation are required as a just transition is not granted under a decarbonization scenario

Supporting Information:

Supporting Information may be found in the online version of this article.

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Citation:

Gayo, E. M., Muñoz, A. A., Maldonado, A., Lavergne, C., Francois, J. P., Rodríguez, D., et al. (2022). A cross-cutting approach for relating Anthropocene, environmental injustice and sacrifice zones. *Earth's Future*, *10*, e2021EF002217. https://doi.org/10.1029/2021EF002217

Received 19 MAY 2021
Accepted 10 APR 2022

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Abstract

The Anthropocene is an uneven phenomenon. Accelerated shifts in the functioning of the Earth System are mainly driven by the production and consumption of wealthy economies. Social, environmental and health costs of such industrialization, however, bear on low-income communities inhabiting severely degraded territories by polluting activities (i.e., sacrifice zones). How global, national and local socio-economic and governance processes have interacted in perpetuating socio-environmental inequalities in these territories has been rarely explored. Here, we develop an historical quantitative approach integrating a novel chemostratigraphic record, data on policy making, and socio-economic trends to evaluate the feedback relationship between environmental injustice and Anthropocene in sacrifice zones. We specifically outline a case study for the Puchuncaví valley -one of the most emblematic sacrifice zones from Chile-. We verify an ever-growing burden of heavy metals and metalloids over the past five decades paced by the staggering expansion of local industrial activities, which has ultimately been spurred by national and transnational market forces. Local poverty levels have declined concomitantly, but this path toward social equality is marginal as costs of pollution have grown through time. Indeed, national and international pollution control actions appear insufficient in mitigating the cumulative impact brought by highly toxic elements. Thus, our sub-decadal reconstruction for pollution trends over the past 136 years from a sediment record, emerges as a science-based tool for informing the discussion on Anthropocene governance. Furthermore, it helps to advance in the assessment of environmental inequality in societal models that prioritize economic growth to the detriment of socio-environmental security.

Plain Language Summary

Costs of the sustained industrialization growth typically bear on low-income communities (i.e., sacrifice zones). In this work, we designed a case study for the Puchuncaví sacrifice zone (Chile) to understand how socio-economic and policymaking processes interact in perpetuating environmental inequality in these territories. Specifically, we integrate data obtained from a new sediment archive for historical pollution, socio-economic trends and environmental policies to evaluate the relationship between environmental injustice and Anthropocene. We observe an ever-growing load of heavy metals and metalloids over the past five decades paced by the staggering growth of local industrial activities, which has ultimately been spurred by national and transnational industrial and economic demands. Poverty levels in local communities declined concomitantly, but such reduction in social inequality is marginal and deceiving as effects of pollution have grown through time. Pollution control actions appear insufficient in mitigating cumulative and emergent impacts of highly toxic elements. Thus, our 136-year reconstruction of the local pollution trajectory serves as a science-based tool for informing the discussion on governance in the

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Methodology: Eugenia M. Gayo, Ariel A. Muñoz, Antonio Maldonado
Project Administration: Eugenia M. Gayo, Ariel A. Muñoz
Resources: Eugenia M. Gayo, Ariel A. Muñoz, Antonio Maldonado
Software: Eugenia M. Gayo
Supervision: Eugenia M. Gayo
Validation: Eugenia M. Gayo, Ariel A. Muñoz, Céline Lavergne, Laura Gallardo
Visualization: Eugenia M. Gayo
Writing – original draft: Eugenia M. Gayo
Writing – review & editing: Eugenia M. Gayo, Ariel A. Muñoz, Antonio Maldonado, Céline Lavergne, Jean Pierre Francois, Delia Rodríguez, Marcelo Mena-Carrasco, Anahí Urquiza, Laura Gallardo

Anthropocene. Particularly, it helps to advance in the assessment of environmental inequality in societal models that prioritize economic growth to the detriment of the ecosystem and social security.

1. Introduction

Human activities have become the prevalent force of change in the Earth System. Accelerated shifts in biogeophysical processes have occurred unequivocally over the last 250 years paced by the rapid and sustained global socio-economic growth experienced globally since the Industrial Revolution, particularly since the Great Acceleration in the mid-twentieth century (Steffen et al., 2015). However, it is still debated whether this human-dominated state should be formally recognized as a new geological time unit: the Anthropocene. Much of this debate is centered on chronostratigraphic markers (Zalasiewicz et al., 2021). Moreover, the Anthropocene concept is keenly contested as it evokes and foists generic agencies upon the entire humanity. The fact that the production and consumption of countries within the Organization for Economic Cooperation and Development (OECD) have contributed mostly to post-industrial transformations (Steffen et al., 2015), reveals that the Anthropocene is far from being an even phenomenon. Conversely, it appears structured by political and economic interests of a relatively small group of the global population that lead to imbalanced impacts on socio-ecological systems (Keys et al., 2019; Malm & Hornborg, 2014; Moore, 2017; Pulido, 2019).

Such asymmetries are especially evident in the so-called sacrifice zones (*sensu* Lerner, 2010) that have emerged worldwide at least since the 1950s decade (Dionne, 2016). These represent dangerously degraded territories resulting from heavily polluting activities. Here, environmental, social and health costs of such industrialization bear typically on low-income or minority communities, which is maintained by decisions aimed at increasing consumption and production rates (Mohai & Saha, 2015; Pulido, 2019). Thus, sacrifice zones are deemed as spaces of the Anthropocene (Harrison, 2019; Tironi et al., 2018), where high pollution burdens arise from the interplay between global, national and local socio-economic processes that, ultimately, perpetuate socio-environmental conflicts and hazards. Thus, sacrifice zones are also territories where environmental injustice (or environmental inequality) persists over time, which operationally refers to the disproportionate and uneven pollution burden shouldered by a particular social group (Pellow, 2000). In fact, inhabitants of sacrifice zones do not possess the means to avoid such intense pollution burden and afford to move to cleaner and healthier territories, thus being truly trapped populations (i.e., fenceline communities, Lerner, 2010).

Sacrifice zones appear particularly vulnerable to cross-scale and socio-ecologically complex hazards derived from human-driven changes in biophysical processes (i.e., Anthropocene risks; Keys et al., 2019). The lack of effective environmental governance at local, national and transnational levels might trigger unexpected interactions related to environmental inequality that exacerbate the impact of these emerging systemic hazards, turning governance even more challenging (Keys et al., 2019). Historical perspectives on dynamic interactions among anthropogenic transformations, environmental injustice and decision making might help inform the design of adaptation and mitigation strategies to face present and future Anthropocene risks. Yet, the convergence of these factors in sacrifice zones has been conventionally addressed in the Northern Hemisphere through environmental narratives -i.e., narrations of individuals' experience on losses or gains in socio-environmental well-being brought about by environmental changes-. These narratives have been instrumental for engaging participants in environmental movements as well as shaping the public opinion on socio-environmental conflicts. Nevertheless, available experiences indicate that the explicit integration of environmental narratives and interdisciplinary scientific evidence represents a key step to achieve successful mitigation and adaptation actions (Kelly et al., 2014; Malena-Chan, 2019; Manoiu et al., 2016).

By adopting cross-cutting frameworks from social sciences and geosciences, we evaluate how feedback relationships between pollution, social inequality, environmental regulations and socio-economic trends have operated and evolved in sacrifice zones. Specifically, we outline a case study for the socio-environmental history for the Puchuncaví valley (Figure 1) -one the most emblematic sacrifice zones in Chile-. Thus, relevant data for socio-economic trends were coupled to novel geochemical data from the first long-term proxy record for local environmental pollution trajectory over the last century. We focus on reconstructing temporal variations in the accumulation of heavy metals and metalloids (hereafter, jointly referred as metal(oid)s) in a sediment record (i.e., PUCH0112-AT1 core) as these chemostratigraphic archives are key to track long-term trajectories for heavily polluting industrial activities. More importantly, most metal(loid)s emitted by industrial activities (e.g., copper,

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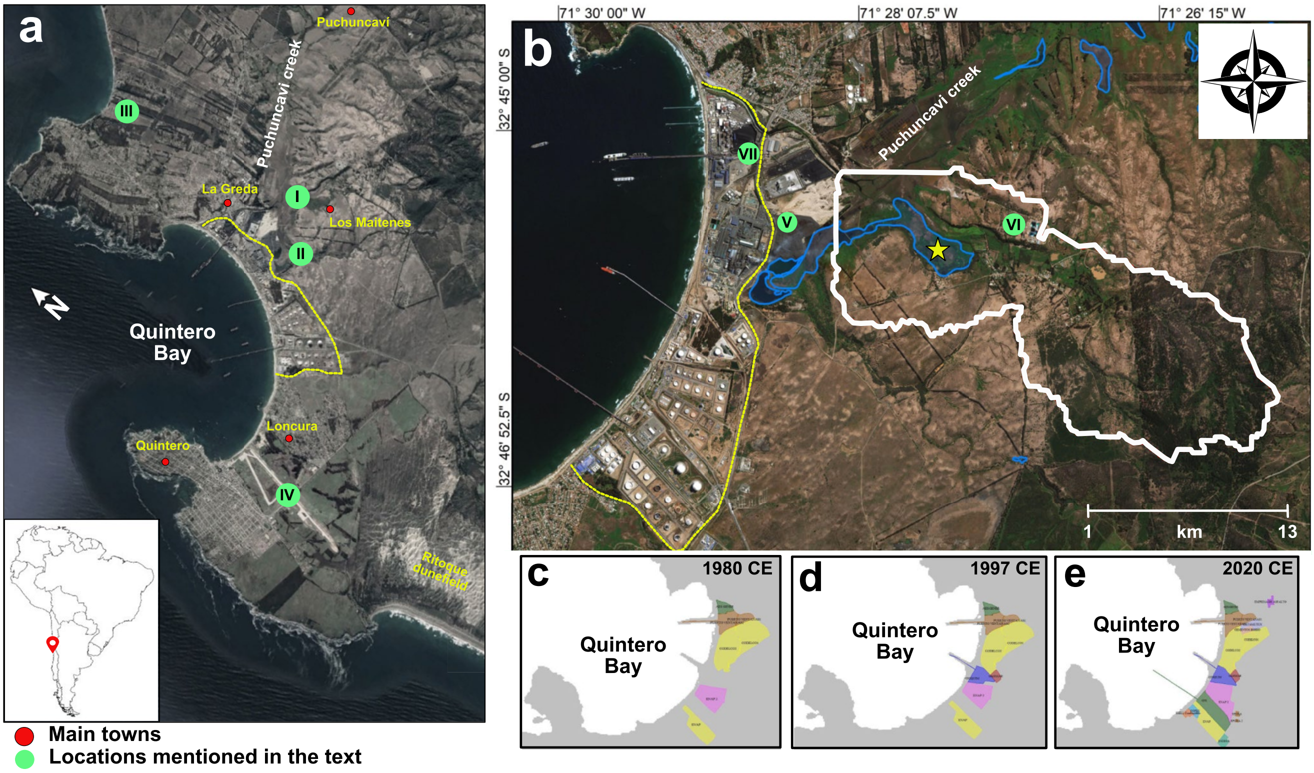

Figure 1. Study area. (a) The yellow line describes the extension for the Ventanas Industrial Area (VIA). Locations mentioned in the text: (I) Los Maitenes and (II) Campiche wetlands, (III) Costa Quilén dendrochronological sampling site (Muñoz et al., 2019), (IV) Quintero Air Base. (b) Details for the study area showing the sampling site for the PUCH0112-AT1 sediment core (yellow star) within the Los Maitenes-Campiche sub-basin (white solid line), (V) the active pile of smelting-slag from the Fundición Ventanas, (VI) the abandoned Los Maitenes tailing dam of copper-sulfide, (VII) coal pile from the AES Gener power plant. (c)-(d) Satellite estimates for the extension of VIA at years 1980, 1997 and 2020 CE.

mercury, lead, arsenic, chromium) remain stocked and, practically, unremovable from ecosystems by natural processes (Förstner & Wittmann, 1981), thus leading to cumulative stress -in turn to persistent hazards-to environment and human health.

Aside from the advance that this study could represent for understanding the long-term configuration of sacrifice zones in the Southern Hemisphere, we propose an interdisciplinary approach to quantitatively evaluate close links between environmental injustice and Anthropocene. In this context, we understand the Anthropocene not only as a geological epoch but as the aggregated effects of human activities on the Earth System that are altering its functioning (Gibbard et al., 2021) and are impacting human society in an unbalanced manner (Keys et al., 2019; Rockström et al., 2021). We consider that our integration exercise could lead to a reconsideration of contemporary socio-environmental exclusions as regards intrinsic and extrinsic aspects for one of the most important stressors of socio-ecological systems.

2. Physiography of Puchuncaví Sacrifice Zone

Chile, an OECD country, has exhibited rapid economic growth in comparison to the Latin American region by a 10-fold increase of its Gross Domestic Product (GDP) between 1960 and 2019 CE. Much of the "Miracle of Chile" relies on copper production and export, which led to rapid urban and industrial expansion since the 1960s (Rodríguez-Weber, 2017). Such escalating socio-economic growth patterns have brought severe socio-environmental conflicts in several areas of Chile (Carranza et al., 2020; Delamaza et al., 2017). This is the case of our studied area that hosts metal smelting, coal-fired power plants, port infrastructure as well as fuel and lubricant and gas storage.

The concepts of sacrifice zone and environmental injustice are not assimilated in the Chilean law, as these allude to the act of violating the right to live in a pollution-free environment, which is declared by the current Constitution (Hervé, 2015). Nevertheless, notions of both terms have been increasingly used in the public discourse in Chile over the last decade (Valenzuela-Fuentes et al., 2021; Valenzuela-Perez, 2016). Sacrifice zones in Chile usually refer to territories where power plants and other hazardous industrial activities occur, leading to acute and chronic environmental and health impacts on districts in that residential and industrial zoning areas are immediately adjacent or even mixed (Castan-Broto & Sanzana-Calvet, 2020). The concept of environmental injustice is embraced to demand a safe, clean and healthy environment by grassroots (Valenzuela-Fuentes et al., 2021). Even though the National Institute for Human Rights identifies more than 100 socio-environmental conflicts in Chile (https://mapaconflictos.indh.cl), only five territories are currently recognized in the public discourse as sacrifice zones with the Puchuncaví valley being an iconic case (32°S, Figure 1).

Located in the semi-arid coast of central Chile, this territory has been subject to considerable transformations enforced by economic and political decisions that sustained the take-off of Chile's GDP. Precisely, under a discourse of local, regional and national economic prosperity, the construction of Ventanas Industrial Area (hereafter VIA) began in 1960 CE along the Quintero bay (Figure 1), which represents the first and biggest industrial complex in Chile. Favorable economic and logistical feasibilities made VIA viable here, but also because the valley had land available for future expansion and met ventilation conditions that would reduce pollution damages on an area inhabited sparsely by poor communities (>14,900 inhabitants; DEC, 1964), and historically dedicated to artisanal fisheries and farming (Folchi, 2006). Still, mineral dust and smelting byproducts started to spread over the local population when the state-owned Ventanas copper smelter (using reverberatory furnaces) and a 114 MW coal fired power plant (Ventanas1) came online in 1964 CE, and large open bulk storages of copper concentrates and coal began to accumulate. Since 1977 CE industrial activities have expanded and diversified (Figure 1c), and atmospheric emissions of diverse pollutants have become evident, including particulate matter (PM), nitrogen oxides (NO_x), sulfur dioxide (SO_2) and heavy metals. Much of VIA sources had almost non-existing emission controls until 1998 CE, but a pollution attainment program was established in 1991 CE. Despite the implementation of several actions for controlling the pollution load in VIA, chronic exposure of communities have brought detrimental health effects on local communities, increasing the risks of cardiovascular and respiratory mortality as well as cancer rates (Berasaluce et al., 2019; Salmani-Ghabeshi et al., 2016; Tapia-Gatica et al., 2020; Tume et al., 2019). More importantly, health hazards persist for >51,000 inhabitants as acute massive intoxication events by SO_2 emission spikes, and volatile organic compounds (VOCs) due to poorly controlled fuel transfer have occurred during the recent decade (e.g., http://hdl.handle.net/11250/2596936; https://www.wired.co.uk/article/chile-quintero-pollution). This together with other socio-environmental costs brought about by local industrial activities have led to grassroots actions around issues of "unequal protection" and "to be sacrificed" (Valenzuela-Fuentes et al., 2021; Veas-Basso & Fuentes-Pereira, 2020).

Air quality monitoring data for the Puchuncaví valley goes back to 1997 CE, and it includes continuous measurements of coarse and fine PM (i.e., PM_{10}, $PM_{2.5}$), SO_2, NO_x (see https://sinca.mma.gob.cl/), and sporadic analyses of heavy metals such as arsenic, lead and copper (see https://mma.gob.cl/valparaiso). This hinders devising the socio-environmental history of this sacrifice zone before 1997 CE, but it also falls short for evaluating the efficacy either of environmental regulations, restoration plans or palliative actions. Localized wetlands along the watershed, however, provide ideal depositional environments that allow the development of sediment records that have archived sequentially past metal(loid)s releases by industrial activities. Thus, these paleopollution records have the potential for informing on the air quality even through the pre-industrial period in the Quintero bay (i.e., prior the 1960 decade). Actually, within the VIA, Los Maitenes and Campiche wetlands (Figure 1b) operate as an integrated hydrological, sedimentary and ecological system. Declared as a protected area in 1998 CE due to its high biodiversity significance, this wetland complex (628 ha) has witnessed significant loss and degradation from the increasing growth of the VIA (Figures 1b and 1c), but also from the expansion of urban areas and exotic forest plantations (Badal, 2014; Fernandez, 2011).

3. Data and Methods

3.1. Coring Site, Chronology, Geochemical and Elemental Analyses

We recovered a sediment core (PUCH0112-AT1) from the deepest section of the Los Maitenes wetland using a Livingstone piston-corer (Figure 1). This core site lies ~2 km east from the VIA, practically adjacent to an

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abandoned tailing dam (Minera Los Maitenes) and the massive active pile of smelting-slag from the Ventanas smelter-refinery (Figure 1b). Detailed procedures for the recovery, sampling, chronology, age-depth model, geochemical and elemental analyses for the PUCH0112-AT1 core are presented in the Supporting Information S1 (Text S1 in Supporting Information S1).

We used the ^{210}Pb (lead-210; half-life = 22.3 years) dating method to establish the chronology of PUCH0112-AT1 sediments. This radiometric method is widely used to generate recent chronologies (100–150 years) in lacustrine and marine environments, which relies on estimating the ^{210}Pb activity along the sediment profile -i.e., number of decays per second per Kg (Bq kg^{-1}). Hence, we determined indirectly the total ^{210}Pb activity ($^{210}Pb_{total}$) in sediments by measuring the decay of progeny radionuclide ^{210}Po (polonium) through alpha spectrometry. Because the $^{210}Pb_{total}$ profile reached a nearly constant value in the lower core sections (Figure S1 in Supporting Information S1), we estimated the supported ^{210}Pb activity ($^{210}Pb_{sup}$) as the mean of the five lowermost sections ($^{210}Pb_{Sup}$ = 15.03 ± 0.83 Bq kg^{-1}). The $^{210}Pb_{sup}$ refers to the ^{210}Pb sourced from in situ disintegration of the precursor radionuclide ^{226}Ra (radium), which enters to Los Maitenes sediments via water flux and discharge. To generate ages for sediments, we estimated the unsupported ^{210}Pb activity ($^{210}Pb_{ex}$) by subtracting the $^{210}Pb_{Sup}$ to the $^{210}Pb_{total.}$ The $^{210}Pb_{ex}$ is the fraction sourced by atmospheric deposition (rainfall or dry fallout), which activity decreases with sediment depth. To convert the obtained $^{210}Pb_{ex}$ activity profile into an age-depth model a Constant Initial Concentration (CIC) model was used (see Text S1 in Supporting Information S1).

By using Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7700 ICP-MS), we determined the concentration of metal(loid)s in bulk sediments sampled at a resolution of 1–3 cm-depth within the PUCH0112-AT1 core. These analyses were conducted at the Arizona Laboratory for Emerging Contaminants (ALEC, University of Arizona, US). Sediments were analyzed specifically for industrial-derived metal(loids) such as copper (Cu), mercury (Hg), silver (Ag), lead (Pb), cadmium (Cd), zinc (Zn), tin (Sn), niobium (Nb), phosphorus (P), molybdenum (Mo), boron (B) and antimonium (Sb). Additionally, we measured concentrations for titanium (Ti) and aluminum (Al), which serve as reference elements in the calculation of pollution indicators (see Section 3.3). To evaluate the potential contribution of diagenetic processes on the post-depositional distribution of elements, we calculated dry bulk (BD), organic (OD), siliciclastic (SD) and carbonate (CD) densities based on data from Loss on Ignition analyses.

To explore convergences and divergences in long-term concentrations trends among elements and between organic and inorganic sediment features (i.e., OD, SD, CD), we performed non-parametric statistical tests (Spearman's rank correlations, Mann-Kendall test) and a Principal Component Analysis (PCA) on natural logged variables. All analyses were done in R. 3.6 (R-Development-Core-Team, 2019).

3.2. Proxy-Record Replicability

We assessed the replicability of our elemental profile as a proxy record for tracking long-term variations in the metal(loid)s burden at VIA. Thus, our results for the content of elements in PUCH0112-AT1 sediments were compared against data from the *Cupressus macrocarpa* dendrochemical series developed at Costa Quilén (Muñoz et al., 2019) ~5 km north-west of our coring site (Figure 1a). Long-term trend comparisons are, however, not straightforward due to differences in temporal extents and resolutions between both records. For comparative purposes data from our sediment core were matched to the time span (1960–2008 CE) and resolution (consecutive 4-year windows) covered by the tree-ring series. That is, concentrations for Cu, Ag, Pb, Cd, Zn, Sb, Mo and Al measured in PUCH0112-AT1 sediments were linearly interpolated between two data points, and then averaged between four consecutive data. To compare dispersions of differently scaled datasets, raw concentrations for elements concurrently measured in both records were standardized into z-scores. Then, we evaluated convergences or divergences between these reconstructed long-term trends for the accumulation of each metal(loid). These relationships were evaluated by implementing *t*-tests to detect regime shifts in weighted Pearson correlation coefficients (Rodionov, 2015), applying the Olkin-Pratt corrector, fitting the Huber weight function on normalized time-series and setting cutoff lengths of two intervals (8 years).

3.3. Temporal Variations in Local Pollution Levels

Because we are interested in evaluating temporal variations either in the presence or intensity of pollution by metal(loids) in the Puchuncaví valley, we computed pollution indicators such as Enrichment Factor (EF) and

Geoaccumulation Index (Igeo). In this manner, we are able to evaluate quantitatively and qualitatively the relative contribution of anthropogenic versus natural sources on the long-term accumulation of elements in PUCH0112-AT1 sediments.

EFs allow identifying positive anomalies (i.e., enrichment) in the accumulation of metal(loid)s in sediments from the Los Maitenes-Campiche wetland complex. These anomalies were detected by comparing the measured concentration for a target metal(loid) against the concentration of a reference element (hereafter, lithogenic element), which its content in the sample derives from the Earth's crust. We used Ti as a lithogenic element for the normalization in EF calculations because it represents a conservative and stable crustal tracer in aquatic systems (Shelley et al., 2015). In practice, EFs for our 12 metal(loid)s at each sampled sediment section were calculated following Equation 1 (Muller, 1969).

$$\mathrm{EF} = \left(\left[\mathrm{X_{sample}}\right] / \left[\mathrm{Ti_{sample}}\right]\right) \,/\, \left([\mathrm{X_{BG}}] / [\mathrm{Ti_{BG}}]\right) \tag{1}$$

Where, $[X_{sample}]$ and $[Ti_{sample}]$ are concentrations for the examined metal(loid) and Ti in the sediment sample, respectively. $[Ti_{BG}]$ and $[X_{BG}]$ indicate background levels that account for natural contents for Ti and the target metal(loid). We estimated local background levels for each metal(loid), which refer to the normal or natural concentration of an element in our studied area expected without the anthropogenic influence. In particular, background levels were obtained by averaging concentrations recorded after the identification and removal of outlier observations through the Median ± 2 Deviation method.

Igeo describes the degree of pollution of sediments (Muller, 1969). This index was calculated for 12 metal(loid)s at each sediment section sampled in the PUCH0112-AT1 core. We applied Equation 2 (Muller, 1969), which relates the concentration in the sediment sample ($[X_{sample}]$) and the calculated local background level for the target metal(loid) (i.e., $[X_{BG}]$) through the Median ± 2 Deviation method.

$$\mathrm{Igeo} = \mathrm{Log}_2\left([\mathrm{X}]_{\mathrm{sample}}/1.5[\mathrm{X}]_{\mathrm{BG}}\right) \tag{2}$$

We followed criteria proposed by Zhang and Liu (2002) and Muller (1969) to classify EF and Igeo values into categories. Thus, EFs <1 indicate minimal enrichment and that an important amount of the metal(loid) in the sediment sample may derive from natural sources. Values above one indicate that the element accumulation is explained by anthropogenic inputs, where EF 1–2 indicate moderately enriched sediments and EFs >5 significantly enriched. Igeo values <1 indicate uncontaminated sediments, and thus a negligible contribution of anthropogenic inputs. Igeo values >1 suggest important anthropogenic influence. That is, Igeo in the ranges 1–3, 3–5 and >5 indicate moderately, heavily, and extremely contaminated sediments, respectively.

Anthropogenic concentrations ($[X_{anthropogenic}]$) were additionally calculated to estimate how much amount (i.e., concentration) of each metaloid at a given sampled sediment section is sourced exclusively by human activities via atmospheric deposition. That is, we quantified the anthropogenic burden or load ($[X_{anthropogenic}]$; μg/g) for individual metal(loid)s by using Equation 3 (Norton & Kahl, 1991). In turn, the expected contribution of natural inputs (i.e., geological or edaphic factors) in the accumulation is offset from the total concentration measured in the sample for each element ($[X_{sample}]$). Again, we used Ti as a lithogenic element to account for natural contributions either in the sample ($[Ti_{sample}]$) or background levels ($[Ti_{BG}]$). Local background levels were obtained considering data for the preindustrial local period (i.e., prior 1964 CE), and were assumed as constant in our calculations.

$$\left[\mathrm{X_{anthropogenic}}\right] = \left[\mathrm{X_{sample}}\right] - \left(\left[\mathrm{Ti_{sample}}\right]^{*} [\mathrm{X_{BG}}] / [\mathrm{Ti_{BG}}]\right) \tag{3}$$

We estimated anthropogenic fluxes (X_{flux}) for each examined metal(loid), which account for the accumulation rate (μg cm^{-2} yr^{-1}) for inputs from anthropogenic sources. We inferred X_{flux} at each sampled sediment section by using Equation 4; where $[X_{anthropogenic}]$ refers to the calculated anthropogenic concentration (Equation 3), BD is the dry bulk density estimated for each core section (Data Set S1), and SR indicates the sedimentation rate yielded by the ^{210}Pb age-depth model.

$$\mathrm{X_{flux}} = \left[\mathrm{X_{anthropogenic}}\right]^{*}(\mathrm{BD})^{*}(\mathrm{SR}) \tag{4}$$

Because the socio-environmental degradation by metal(loid)s does not -necessarily- rely on trends for pollution indicators for individual elements, we calculated a pollution load index (PLI; Tomlinson et al., 1980). The PLI describes the magnitude to which the content for all pollutants measured in a sample exceeds local background levels, providing a comprehensive overview for pollution levels by different metal(loid)s; where PLI <1 indicates no pollution, and values >1 reflect high load. We calculated PLI values at each sampled sediment section by following Equation 5 (Tomlinson et al., 1980). Thus, these were obtained as the product between concentrations factors (CF) estimated for our 12 metal(loid)s according to Equation 6, and then by raising the multiplication result to the exponent of 0.083 (i.e., the quotient between 1 and the total number of elements examined). For individual CF estimates, we considered the metal(loid) concentration measured at each sediment section ($[X_{sample}]$) and the local background concentration ($[X]_{BG}$) inferred through the Median ±2 Deviation method.

$$\mathrm{PLI} = \left(\mathrm{CF}_{X1}{}^{*}\mathrm{CF}_{X2}{}^{*}\mathrm{CF}_{X3}{}^{*}\mathrm{CF}_{X4^{*}}\mathrm{CF}_{X4^{*}}\mathrm{CF}_{X5}\ldots\right)^{0.083} \tag{5}$$

$$\mathrm{CFx} = \left([\mathrm{X}]_{\mathrm{sample}}/[\mathrm{X}]_{\mathrm{BG}}\right) \tag{6}$$

By assembling PLI values obtained at each sampled sediment section, we generated a continuous time-series to identify temporal changes in the local pollution burden over time (i.e., pollution phases). Specifically, we implemented a sequential *t*-test algorithm to detect statistically significant regime shifts (Rodionov, 2004, 2006) in the PLI mean at a cutoff length of five intervals and by fitting an Autoregressive Model by Ordinary Least Squares.

3.4. Socio-Economic Trends and Environmental Regulations

Within the framework of Anthropocene risks, the long-term trajectory of sacrifice zones is determined by the interplay of socio-economic and decision-making processes that operate from local to transnational levels (Keys et al., 2019). To reflect this at the local scale, we considered proxy data for changes in economic activities in the Puchuncaví valley brought directly or indirectly by environmental pollution, zoning planning, displacement of traditional practices and labor reallocation. Thus, we selected a time-series outlined by Badal (2014) for livestock and agricultural Gross Production Value (GPV) and total sown area in the Puchuncaví valley for the period 1964–2007 CE. A time-series for the area growth of VIA throughout the interval 1960–2008 CE was generated by using a change detection method based on the analysis of multi-temporal aerial photographs and LANDSAT TM imagery. We also compiled main infrastructure milestones to capture the long-term industrial diversification in the Quintero bay.

A time-series for the production of copper refined at Ventanas smelter-refinery (Folchi, 2006; Muñoz et al., 2019) allowed tracking the intensity of local industrial activities since 1966 CE. Although this is a relatively small operation (400,000 ton/year) that just accounts for the 7% of the national production (Perez, 2017), its product provides one of most world's purest copper concentrates (~99.9%), which is in high demand globally. Hence, we deem that the time-series for local copper production is concurrently capturing trends in national export income and tax revenues as well as international market forces in terms of investment and demands. Given that much of the industrial activities conducted in VIA are associated to primary and secondary productive sectors (oil refining, chemicals, bulk ports, production of copper, energy and chemical supplies) that have contributed mostly to the economic growth of Chile (Schmidt-Hebbel, 2006), we also considered data for the Chilean Gross Domestic Product (GDP, data.worldbank.org/indicator).

In this work, social inequality is understood as unequal distributions of opportunities (life chances) and conditions (income, goods, wealth) across individuals that belong to a given social group (Guidetti & Rehbein, 2017). We are aware that accounting for these dimensions is challenging as social inequality is determined by structural factors (e.g., redistributive policies, socioeconomic status, access to services, institutions, technologies) as well as group membership (ethnicity, gender, religion). Althought different methodological and theoretical approaches have emerged either in economics or sociology, there is still no consensus on what are the best descriptors for such multivariate phenomena (Guidetti & Rehbein, 2017). However, Brunori et al. (2013) by implementing a cross-country comparison evidence that uneven distributions of opportunities and income are strongly interlinked and feed back into each other, thus hampering intergenerational social and economic mobility. Based on this feedback relationship, here we used the national Gini Index as a proxy for social inequality. Specifically, we considered the smoothed 1960–1984 CE reconstruction for the Gini Index developed by Rodríguez-Weber (2017) on the aggregation of different historical statistical sources for income earner distribution. This set was complemented with estimates of the Gini Index by the World Bank (data.worldbank.org/indicator) for the period 1987–2006 CE as well as with data for the percentage of the national and local population living in poverty (CASEN, 2021). These data account for unidimensional poverty -i.e., in terms of income and the poverty threshold-as official multidimensional measures of poverty in Chile at communal levels have been only recently implemented since 2013 CE. Even so, poverty levels in the country are directly related to social inequality as low incomes lead to unequal distribution of opportunities and capabilities to avoid deprivation (Fernandez-Chicharro, 2016). Actually, the extreme neoliberal economic framework of Chile has, for years, conceived education, health and water as consumption goods, limiting structurally the access to basic and fundamental needs (Harvey, 2007). Thus, economic poverty represents the cornerstone of the process of social exclusion (i.e., social disadvantage) in Chile (Muñoz-Arce & Pantazis, 2018).

To address the relative impact of decision making on the management and control of pollution in the Puchuncaví valley, we constructed a timeline of official regulations (Supreme Decrees) that proliferated in Chile between 1991 and 2002 CE. Because emissions controls from VIA's stationary sources were not specifically targeted in reducing metal(loid)s but PM and SO_2 emissions, we also bring into our comparisons time-series for mean annual [PM_{10}] and [SO_2] measured by the local air quality monitoring network since 1997 CE (Badal, 2014).

4. Results and Discussion

4.1. Metal(loid) Contents in the Sediment Core

The PUCH0112-AT1 sequence spans the last 136 years from 1872 to 2008 CE (Text S1, Figure S1 in Supporting Information S1), affording a sub-decadal reconstruction for the accumulation of metal(loid)s during the last century (Figure 2). Details for age-depth model, sediment stratigraphy and associations between elemental and geochemical variables are provided in the Supporting Information (Text S1–S3, Figures S1–S4, Table S1 in Supporting Information S1). All elements varied vertically along PUCH0112-AT1 sediments (Data Set S1), thus reflecting temporal fluctuations in the environmental burden of metal(loids) since the preindustrial local period to recent decades (Figure 2). Lithogenic elements (Ti and Al) varied among temporal intervals, but with no clear long-term trend (Figure 2) and unrelated to patterns observed for the rest of metal(loid)s (Text S3, Figure S3 in Supporting Information S1). Cu, Hg, Ag, Pb, Cd, Zn, Sn, Nb, P, Mo, B and Sb have followed a definite convergent upward trend (Figure 2) and an interconnected long-term dynamic (Text S3; Figure S3 in Supporting Information S1). Minimum concentrations for these metal(loid)s occur during the interval 1872–1963 CE, and increase steeply since 1967 CE to reach maximum values by the late-1980s and mid-1990s decades (Figure 2). Contents of Pb, Nd, Hg Cu, Mo, Sn, Sb and B in sediments started to decline moderately since 1997 CE.

We found no evidence for the effect of sediment organic and inorganic densities (OD, SD, CD) on the content of elements in our sediment core (Figures S3 and S4, Table S1 in Supporting Information S1). This implies that temporal variations in concentrations could be genuinely linked to changes in atmospheric deposition of metal(loid)s (Text S3 in Supporting Information S1). The association between Cu, Hg, Ag, Pb, Cd, Zn, Sn, Nb, P, Mo, B and Sb (Table S1, Figure S4 in Supporting Information S1) confirm our notion that reconstructed trends for these elements are best explained by releases from stationary and diffuse sources related to local industrial and urban activities (Text S3 in Supporting Information S1). Specifically, from local anthropogenic emissions, including non-ferrous metal smelting, coal fired power generation, traffic, mine tailings as well as active piles of coal and copper concentrates and smelting-slag (Barraza et al., 2017; Montenegro et al., 2009; Muñoz et al., 2019; Parra et al., 2014a; Rueda-Holgado et al., 2016; Salmani-Ghabeshi et al., 2015).

Raw metal(loid) concentrations in the PUCH0112-AT1 sediments are significantly higher by several orders of magnitude from the *C. macrocarpa* dendrochemical series (Table S2 in Supporting Information S1). Physicochemical (differential solubilities, redox conditions), pedogenic and physiological factors (species-specific metal uptake thresholds or mobility of elements) could explain such difference (Cutter & Guyette, 1993; Monticelli et al., 2009; Watmough, 1999).

The difference referred to above could also be partially explained according to the spatial gradient in the dispersion and deposition of elements across the Puchuncaví valley and the Quintero bay. Our PUCH0112-AT1 sediment core was recovered in the area immediately adjacent to the industrial complex, and the Costa Quilén dendrochronological sampling site lies ~6.5 km north-west of the VIA at the northernmost point of the bay (Figure 1a). Air circulation over the valley is complex as it results from the superposition of large-, meso- and

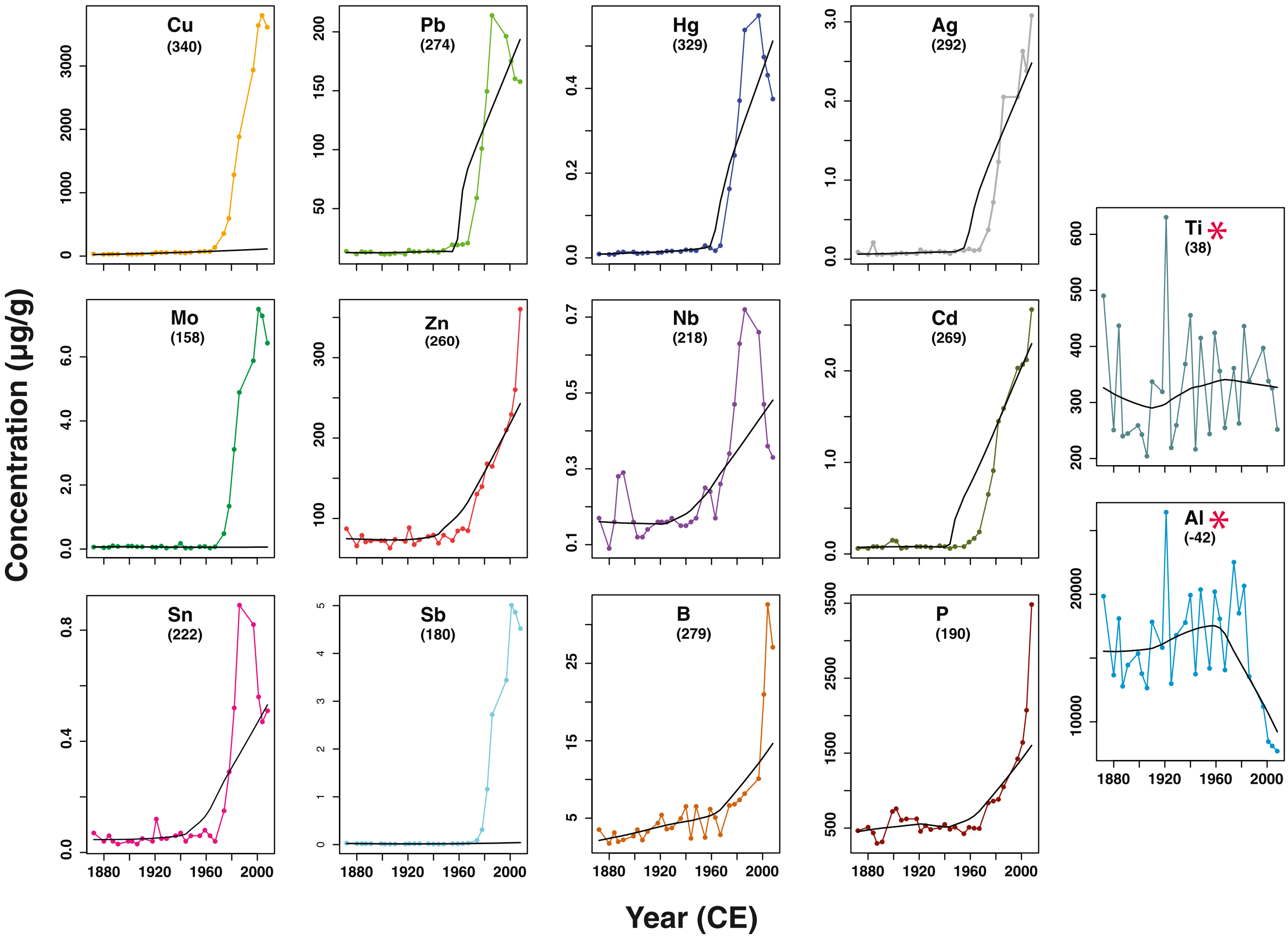

Figure 2. Metal(loid) concentrations (μg/g) in sediments of the PUCH0112-AT1 core for the period 1872–2008 CE. Dark curves describe long-term trends detected from the Mann-Kendall by fitting LOESS smooth curves; note that for Cu, Mo and Sb we applied a bandwidth of 0.8. Numbers in brackets represent Kendall Score values for each element and red asterisks indicate statistically non-significant trends ($p > 0.01$).

local scale dynamics. Thus, while both Los Maitenes wetland and Costa Quilén are affected by VIA, the impact is differentiated by meso and local scale circulation patterns, and their distance to the emission sources. For instance, Costa Quilén is affected by the coastal wind dynamics of the bay to the north. The Los Maitenes wetland is subject to the local wind patterns determined by the contrast between the Quintero bay, and inland along an west-east direction. The combined effect of source-proximity and wind direction has been systematically indicated as the main driver for spatial variations in the content of elements in local rainfalls (Cereceda-Balic et al., 2020), vegetation (Gorena et al., 2020) and topsoils (De Gregori et al., 2003; Parra et al., 2014b; Salmani-Ghabeshi et al., 2015; Tapia-Gatica et al., 2020). Indeed, independent geostatistical models (González et al., 2014; Tapia-Gatica et al., 2020; Tume et al., 2019) consistently reproduce lower concentrations of metals around the northern tip of the Quintero bay and higher concentrations in sites proximal to VIA (e.g., La Greda and Los Maitenes towns, Figure 1a).

Despite differences in absolute concentrations between the dendrochemical record and PUCH0112-AT1 sediments, we verify clear convergences in standardized trends over the period 1964–2008 CE (Figure 3). Sustained increases in Cu, Pb, Ag, Cd, Sb, Mo and Zn since 1964 CE onwards are coherently reproduced, as well as modest reductions in Pb, Sb and Cu during the most recent decade. Statistical analyses (i.e., *t*-tests for correlation shifts) indicate positive and significant relationships between Cu, Sb, Zn, Cd, Mn, Pb and Mo (Figure 3, Table S2 in Supporting Information S1). Al concentrations, however, covaried inversely, likely reflecting inter-site differences in geological (i.e., lithogenic) or edaphic processes and sources. Still, the good agreement between both archives in replicating long-term trends in most elements, confirm that our sediment geochemical profile serves

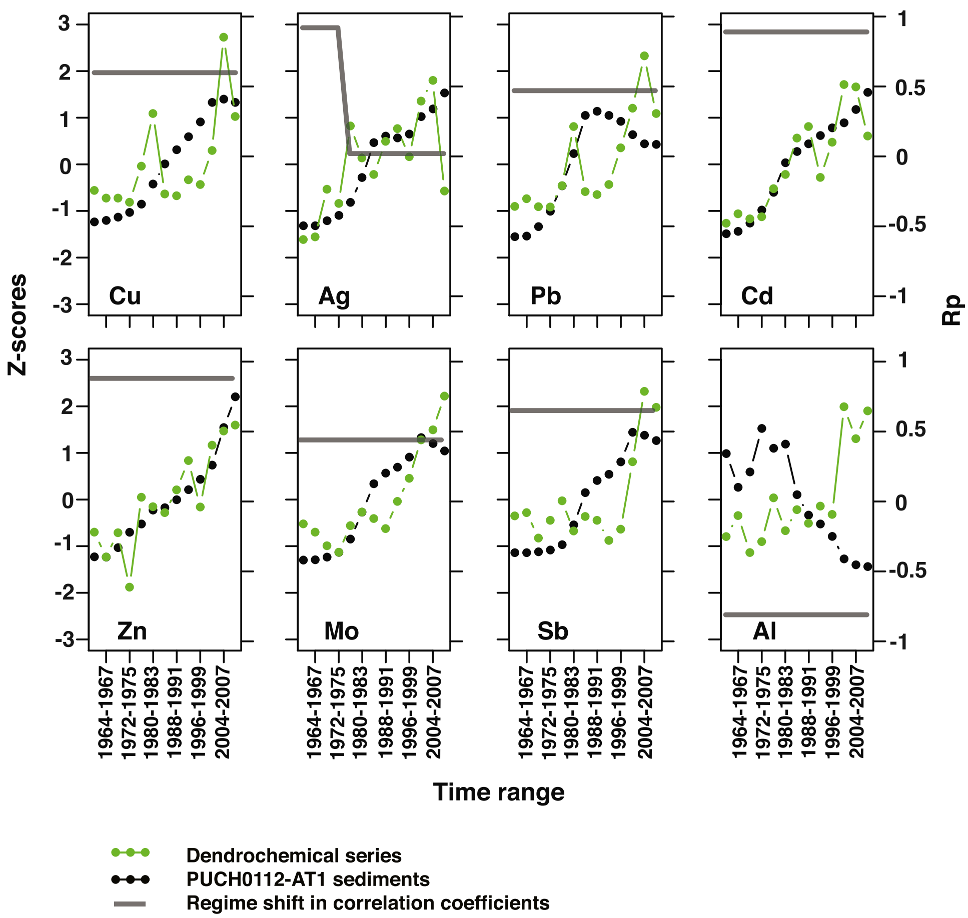

Figure 3. Trend comparisons in standardized concentrations (z-scores) for metal(loid)s shared between the *C. macrocarpa* dendrochemical series (Muñoz et al., 2019) and the PUCH0112-AT1 record. Horizontal grey lines indicate regime shifts in weighted correlation coefficients (Rp) at $p = 0.05$ and cut-off lengths of two intervals (i.e., consecutive 8 years).

as a representative and reliable proxy-record for tracking sub-decadal variations in pollution burden brought by industrial and urban activities in the Puchuncaví valley.

4.2. Socio-Environmental Trajectory for the Puchuncaví Sacrifice Zone

Calculated pollution indicators, anthropogenic concentrations and fluxes consistently point to sub-decadal variations in pollution burden during the past 136 years (Figures 4a and 4b; Data Sets S2–S3). The PLI time-series attest for ever-growing and cumulative impacts of anthropogenic inputs over the last century, describing a long-term trajectory marked by three pollution phases intimately linked to the VIA dynamic (Figure 4c): Phase I (1872–1974 CE), Phase II (1978–1997 CE) and Phase III (2001–2008 CE). These stages are paced by trends in Chilean socio-economic growth, and in turn concomitant increasing demands for production of copper concentrates and energy from fossil-fuel combustion. In fact, the chronology and extent for these phases follow main transitions in the Chilean GDP, copper production in the ENAP Ventanas smelter-refinery and the local capacity for power generation (Figures 4 and 5). The latter is reflected by the increasing number of power plants that have come online at VIA to strategically supply copper production (Rojas, 2015).

The Phase I covers the local pre-industrial period (1872–1960 CE) until 1974 CE (Figure 4c). EFs (<1) and PLI values suggest negligible contribution of anthropogenic inputs up to 1963 CE, and prevailing negative Igeo values attest for uncontaminated sediments (Figures 4a and 4b). Yet, discernible depositions from anthropogenic inputs (i.e., peaks in anthropogenic concentrations; Data Set S3) are evident in some metal(loid)s since 1925 (Cu, Hg, B), 1944 (Ag, B) and 1955 CE (Nd, Cd). These might reflect local emissions from domestic wood burning, which represented the main heating and cooking source for communities that relied on agriculture and artisanal

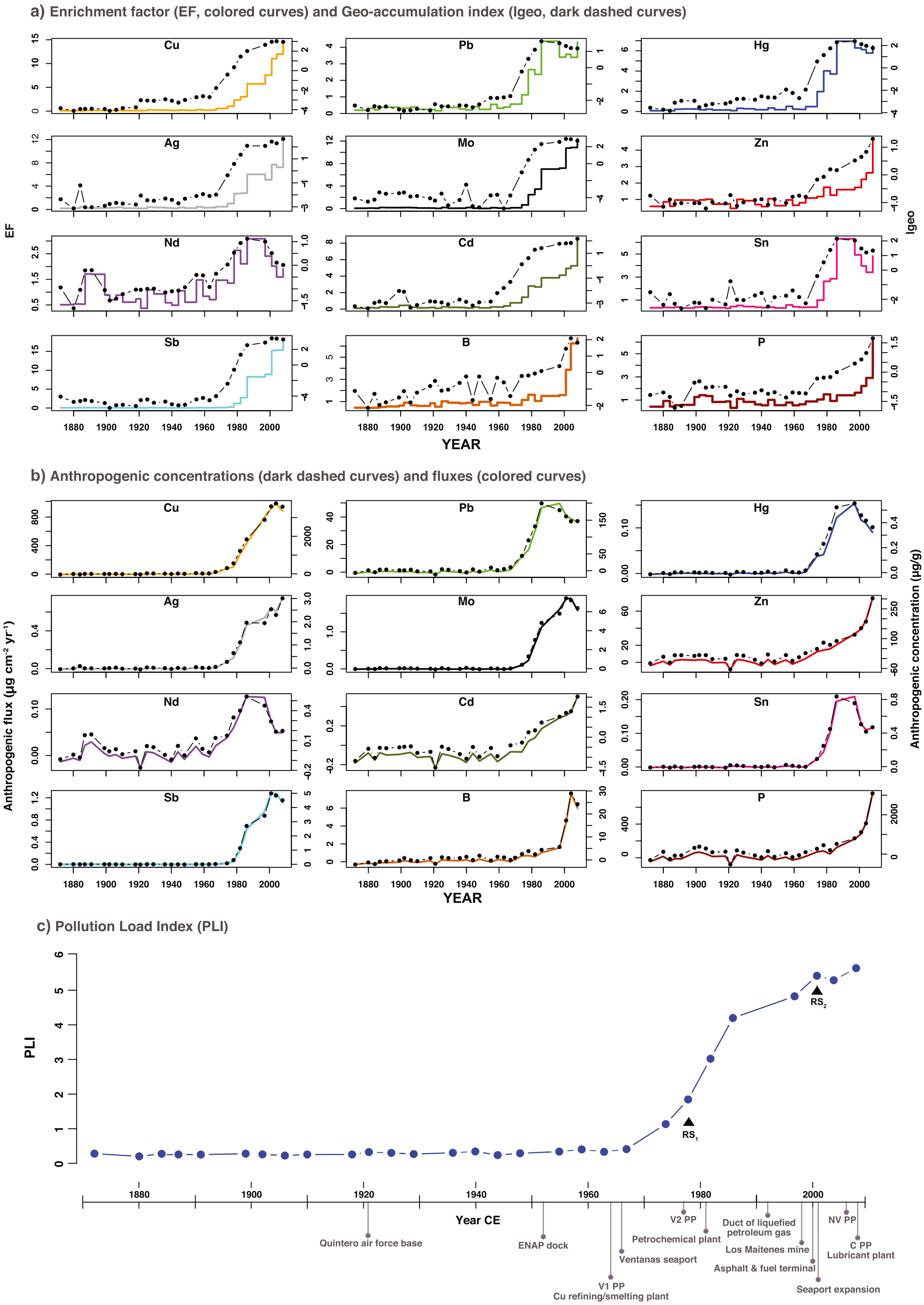

Figure 4. Calculated pollution indicators (a-b) and reconstructed Pollution Load Index (PLI) showing statistically significant regime shifts RS_1 and RS_2 (c). Estimates for anthropogenic fluxes are based on the sedimentation rate of 0.26 cm/year yielded by the age-depth model (see Text S1 in Supporting Information S1). At the bottom, main infrastructure milestones for the Quintero bay. Note that Ventanas1, Ventanas2, Nueva Ventanas and Campiche power plants are abbreviated as V1 PP, V2 PP, NV PP and C PP, respectively.

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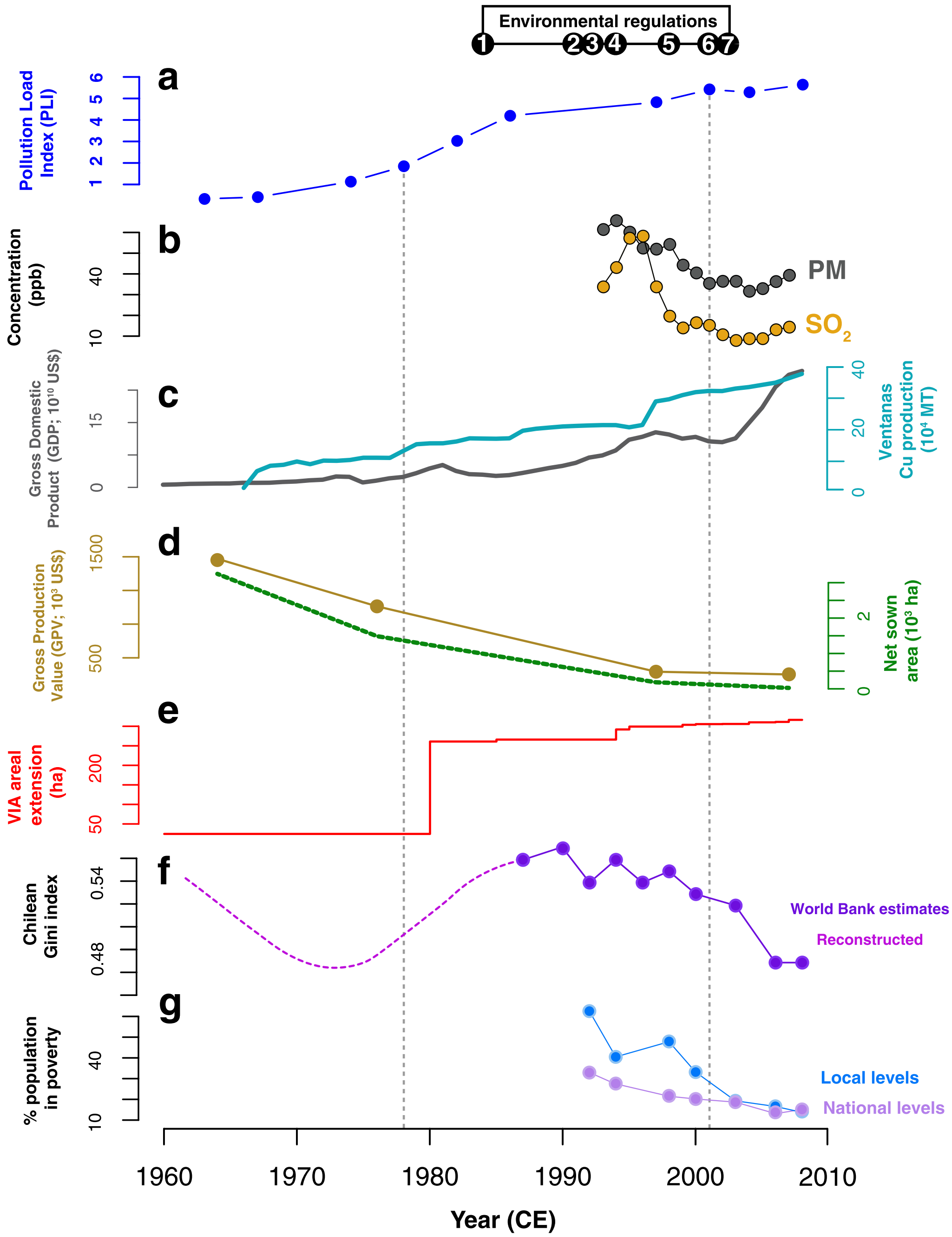

Figure 5. Relationship between historical pollution levels, environmental regulations and socio-economic trends between 1960 and 2008 CE. Vertical grey dashed lines indicate the chronology for statistical significant regimen shifts (RS_1 and RS_2). (a) Reconstructed Pollution Load Index (PLI) against environmental regulations enacted in Chile, where 1: DS 86/1984, 2: DS185/1991, 3: DS 252/1992, 4: DS 346/1994, 5: DS 59/1998 and DS 16/1998, 6: DS 45/2001, 7: DS 113/2002. (b) Measured mean annual concentrations for coarse particulate matter (PM_{10}) and sulfur dioxide (SO_2) in local air quality monitoring stations between 1993 CE and 2007 CE. (c) Annual production of refined copper by the Ventanas smelter-refinery versus the Chilean Gross Domestic Product (GDP). (d) Local livestock/agricultural Gross Production Value (GPV) against the net area sown in the Puchuncaví valley. (e) Estimated area for the VIA growth. (f) Historical reconstruction (Rodríguez-Weber, 2017) and World Bank´s estimates for the Chilean Gini index. (g) Official statistics for percentage of the national and local population living below the poverty line.

fishing in the Quintero bay until the 1960s (Rojas, 2015). Similarly, these could be associated with the incipient infrastructure endowment of the bay during the first half of the twentieth century (Figure 4c). Particularly, to traffic, aviation, and shipping emissions from the Quintero Air Base (Figure 1a) and the ENAP dock. Such discrete incursions in anthropogenic inputs could be also explained by industrial and coal-burning emissions from nearby production centers that flourished since the 1940s decade as part of the Import Substitution Industrialization model launched by the Chilean government to reactivate the national economic growth and increasing copper exports (Del Pozo, 1989; Rodríguez-Weber, 2017). In fact, Los Maitenes-Campiche complex has probably received inputs from the wind-dispersion of airborne particles and gases released by other sources from central Chile. Particularly from Chagres and Caletones copper smelters (Gidhagen et al., 2002; Olivares et al., 2002), which started operation in 1832 and 1912 CE, respectively.

Clear evidence for the impact of local industrial activities in PUCH0112-AT1 sediments manifested around 1967 CE. The contribution of anthropogenic inputs increased steeply shortly after the Ventanas1 coal-fired power plant and the smelter-refinery came online at VIA (Figure 4b). The contribution of anthropogenic sources for Cu, Hg, B, Ag and Cd starts to equal natural loads since 1967 CE (Data Set S3). Pollution indicators and PLI coherently increased (Figures 4a and 4b), but negative Igeo values and EFs <1 suggest that the first decade of energy and copper production at VIA led to a relatively low pollution burden in the Puchuncaví valley.

The initial VIA development played an important role in doubling the Chilean production of refined copper (Folchi, 2006; Ulloa et al., 2017) and in raising the contribution of this industry to the national economic growth (Schmidt-Hebbel, 2006). This is, in turn, reflected in the sustained increase in the national GDP (Figure 5c) and a downward trend in the national Gini Index (Figure 5f), which appears especially important in relation to levels reconstructed from 1910 to 1950 CE (Gini Index >0.50; Rodríguez-Weber, 2017). This fall in inequality arises from state reforms that promoted since the 1950s decade to regulate economic activities and income distribution, and thus to reduce the control that elites had over the power and wealth distribution since the nineteenth century. In particular, such structural change was achieved by raising wages and promoting the migration of highly vulnerable agrarian populations to urban areas (Rodríguez-Weber, 2017). In the Puchuncaví valley, the rural flight and concomitant loss of agriculture work force throughout the 1960s decade was comparatively higher than in other nearby rural areas (Sabatini et al., 1995). This has led to propose that such loss of rural population is not solely explained by state-driven structural changes, but could also be linked to the progressive environmental degradation caused by emissions from the Ventanas smelter-refinery (Badal, 2014; Folchi, 2006; Sabatini et al., 1995). This latter is consistent with the increased contribution of anthropogenic inputs for metal(loids) recorded in PUCH0112-AT1 sediments since 1967 CE (Figure 4).

Independent *C. macrocarpa* dendrochemical series for Costa Quilén and two control sites at Isla Negra ~80 km south of VIA, indicate that upward trends in the accumulation of several metal(loid)s observed in the valley from 1964 CE onwards is an idiosyncratic phenomenon not noticeable in nearby coastal towns (Muñoz et al., 2019). A reconstruction for the long-term Cu accumulation in two lakes adjacent to the El Teniente mining and refinery operation (190 km SW of VIA; von Gunten et al., 2009) suggest an equivalent pollution burden over Andean "company towns" (Caletones, Sewell) maintained by the North American Braden Corporation till 1970 CE. Beyond these geochemical comparisons for contamination degrees among territories, different lines of evidence indicate that the incipient pollution burden brought about by the initial operation of VIA started to contribute to environmental inequality in the Puchuncaví valley. In fact, local communities mobilized in 1966 CE to denounce adverse impacts of toxic emissions on human health and crop yields (Sabatini et al., 1995). Time-series for local Gross Production Value (GPV) and net sown area (Figure 5d) indeed attest for concurrent declines in livestock and agriculture production (Badal, 2014; Folchi, 2006). Official data for local income are inexistent for this period, but the dramatic drop in the GPV (Figure 5d) coupled to the loss of arable land and agriculture workforce (Badal, 2014; Folchi, 2006) likely led to Puchuncaví communities going opposite to the national trend in terms of social inequality. Labor reallocation to the industrial sector was minimal (Chahuan, 2019), so the displacement of traditional economies -including fisheries- and the reduction of their profits after VIA started operation might have increased local poverty and exclusion levels.

The second pollution phase is marked by a major regime shift in the PLI at 1978 CE ($RS_1 = 3.2$, $p < 0.05$; Figure 4c). This matches to the start-up of Ventanas2 power plant (220 MW), increased capacities for copper annual production at VIA and the greatest expansion of these industrial facilities through a major reform (DS 86/1984) that relaxed the land-use regulation for the Quintero bay (Figures 4 and 5). EF (>2) and Igeo (>+1) values boosted between 1986 and 1997 CE insofar as residential and industrial zoning limits and manufacturing types were extended (Figure 4). Indeed, relative contributions of anthropogenic inputs increased abruptly at 1986 CE, and anthropogenic fluxes raised at an accelerated pace. This is especially true for Cu, Hg, Cd, Pb, Ag, Mo, Sn and Sb, whose anthropogenic sources account for >93% of the deposition (Figure 4b).

The ever-growing pollution load during the period 1978–1997 CE persisted although three environmental regulations (DS185/1991, DS 252/1992, DS 346/1994) that were in force to control PM, As and SO_2 emissions from the smelter-refinery and both power plants (Figures 5a and 5b). During this period, the production of copper concentrates rose rapidly either at national-scale or VIA (Figure 5c) as foreign capitals and demands increased (Schmidt-Hebbel, 2006; Ulloa et al., 2017). The Chilean GDP boosted accordingly (Figure 5c), but such unprecedented economic growth was accompanied by a historical increase in income inequality by 1978–1990 CE (Figure 5f; Rodríguez-Weber, 2017). This counterintuitive relationship between income distribution and economic growth for the period 1978–1997 CE is intimately linked to the political and economic scenario that characterized Chile during this period. Specifically, rooted in radical socio-economic transformations that the right-wing military dictatorship of Augusto Pinochet (1973–1990) imposed through a neoliberal model that prioritized the growth strategy based on mining and agricultural exports (Harvey, 2007). Hence, by repressing unions and political parties, reducing the social expenditure, promoting financial liberalization, and privatizing state companies, the elite was again favored and recovered the distributive power (Rodríguez-Weber, 2017). Such elite-biased distributive policies coupled to the environmental degradation and the sustained drop either in farming production (net sown area) or profits (GPV Figure 5d), possibly worsened income inequality and wellbeing among Puchuncaví communities since the late 1970s.

As the democracy returned in 1990 CE and a center-left coalition began to govern, Gini Index as well as national and local poverty levels started to fall progressively (Figure 5g) due to the implementation of more progressive redistribution policies (Rodríguez-Weber, 2017). Although this downward trend can be attributed to increased economic growth and social spending (Rojas-Vallejos, 2018), the rapid reduction in the percentage of the population living below the poverty line in the Puchuncaví valley could be also explained by the re-orientation of the local economy to services for industry and tourism (Chahuan, 2019). Still, if there is a relationship between Chilean economic growth and decreasing local poverty levels, this does not eliminate social inequality in the area. Conversely, data compared here (Figures 5c–5g) suggest that the "shared prosperity" principle embraced by the World Bank Group (2015) has not operated in the territory. Poverty levels in Puchuncaví have been indeed comparatively higher than national levels until 2003 CE (Figure 5g). Sporadic data for other inequality indicators -e.g., indexes for social inclusion (Cáceres-Seguel et al., 2020) and human development (MIDEPLAN, 2005)- show that the lack of ambitious redistributive policies have maintained local communities under higher inequality levels than other municipalities in the country. This, together with the fact that inhabitants of this territory have experienced an ever-increasing pollution burden, implies that the access to better living standards and well-being has systematically differentiated. Thus, we identify environmental injustice as Puchuncaví communities have disproportionately suffered adverse impacts of industrialization, while other Chilean communities have benefited from those, which is also expressed in poverty and exclusion, which has remained apparent since at least 1967 CE.

Pollution burden by metal(loid)s kept increasing over the 2000 decade with PLI values ranging from 5.2 to 5.4 between 2001 and 2008 CE (Figure 4c). This trend leads to another regime shift in the PLI at 2001 CE ($RS_2 = 0.73$, Figure 4c). This pollution Phase III (2001–2008 CE) arose from the staggering growth in the national economy and VIA production capacities achieved by the expansion of the seaport, two additional coal-fed power stations as well as the installation of a lubricant manufacturing plant (Figure 4c). EF (>10.5) and Igeo (~3) values for Cu, Ag, Mo and Sb increased steadily up to lead to heavy enrichment and contamination of sediments dated at 2001–2008 CE (Figures 4a and 4b). Anthropogenic fluxes prevail over natural inputs, and >66% of the element burden in sediments is merely explained by emissions from anthropogenic activities at VIA (Figure 4b). This pattern is particularly pronounced for P, Ag, Cd and Zn, which started to duplicate anthropogenic concentrations almost synchronous to the opening of the lubricant plant and the Nueva Ventanas power station (Figures 4b and 4c). Compared to the long-term *C. macrocarpa* dendrochemical for Isla Negra (Muñoz et al., 2019) the sustained increase in the pollution burden in the Puchuncaví territory appears again as a distinct phenomenon. Spatial characterization for metal(loid) concentrations in recent topsoils either from locations adjacent to VIA or nearby non-industrialized townships confirm this pattern (Parra et al., 2014a; Salmani-Ghabeshi et al., 2015). The territory close to the industrial complex displays the highest concentrations in toxic elements (e.g., Cu, As, Pb), bringing to unacceptable levels of carcinogen risk in children (Salmani-Ghabeshi et al., 2016; Tapia-Gatica et al., 2020), and increased chance of epigenic DNA changes (i.e., methylation) in the adult population (Madrid et al., 2022). Furthermore, the Puchuncaví valley shows increased deterioration measured as the ecological risk index (Tapia-Gatica et al., 2020) and with adverse impacts on marine ecosystem health (Oyarzo-Miranda et al., 2020). More importantly, De Gregori et al. (2003) show that the magnitude and amplitude for the pollution burden over this territory is considerably higher compared to other national industrial areas where mining or metallurgy operations exist.

During Phase III, the national pollution attainment program was strengthened with the enactment of three additional regulatory instruments (DS 59/1998, DS 45/2001, DS 113/2002, Figure 5a) that put in place emission standards for SO_2 and PM from power plants and copper smelter-refineries. These standards indeed led to a rapid and sustained reduction in mean annual concentrations for PM_{10} and SO_2 since 1998 CE as measured in air quality monitoring stations from the Puchuncaví valley (Figure 5b). Nevertheless, our reconstructed PLI does not follow the downward trend recorded locally for PM_{10} and SO_2 during the 2000 decade. This indicates that the pollution burden by metal(loid)s cannot be solely explained by inputs from the smelter-refinery and local power plants, but that the chronic degradation of this territory is also brought about by inputs from nearby industrial stockpiles, urban activities and other stationary sources.

Still, we verify that the six environmental regulations enacted in Chile since the 1990 decade (Figure 5a) had some impact on metal(loid) emissions at VIA. Actually, moderate decreases in pollution indicators for Hg, Pb, Cu, Mo, Sb, Nb and Sn occur just after 1998 CE (Figures 4a and 4b). Synchronized drops in these elements are replicated in other paleopollution records from South America (Cooke et al., 2009; 2011; De Vleeschouwer et al., 2014; Eichler et al., 2015, 2017) and elsewhere (Marx et al., 2016; Schuster et al., 2002; Schwanck et al., 2016). This implies that national and international actions to reduce metal(loid) emissions might have contributed to the lowering of these elements in our study area. Global inventories point to important declines in Hg and Sb emissions over the period 1990–2010 CE due to the mercury phase-out from commercial products (Zhang et al., 2016) or the introduction of non-Sb brake pads in vehicles (Tian et al., 2014). Also, traffic lead emissions have started to decline after the unleaded gasoline entered the Chilean market in 1993 CE, but specifically after it was mandated in 1998 CE (DS 16/1998; Figure 5a).

Our elemental analyses of PUCH0112-AT1 sediments do not allow assessing relative contributions of seven environmental policy instruments versus other global and national actions in reducing locally anthropogenic burdens over the 2000 decade. Yet, the impacts of this policy package appear lagged in relation to its launch in 1991 CE (DS185/1991; Figure 5a). We rule out that this pattern reflects a quasi-decadal delayed signal in element accumulation in Los Maitenes wetland due to slow fluxes from the water column to surface sediments, as metal adsorption and co-precipitation in wetlands is an almost instantaneous process (Matagi et al., 1998). This lag is also replicated in the Costa Quilén *C. macrocarpa* dendrochemical series (Muñoz et al., 2019), so it seems associated with other factors rather than depositional processes.

We suspect that this pattern most likely follows from the "graduality principle" in the Chilean environmental legislation, which deals with the trade-off between the need to decontaminate and the magnitude of the capital to be invested for implementing mitigation measures. In our case, the decontamination action launched in 1991 CE involved a voluntary agreement that fixed stepwise implementations until 1996 CE, but just achieved in 1999 CE (Folchi, 2006). Even so, Hg, Pb, Cu, Mo, Sb, Nb and Sn reductions appear to come out as co-benefits from measures that regulate PM and SO_2 emissions from coal-burning and copper production. Thus, the lowering in anthropogenic inputs for these elements might in part arise from the introduction of baghouse filters for particulate removal in VIA power plants. Covariations between Sb, Cu and Mo are expected as tetrahedrite and molybdenum-rich porphyries have been traditionally processed in the Ventanas smelter-refinery (COCHILCO, 2016). So, the convergent bust might be also related to the replacement of oxygen-injection reverberatory furnaces by electrolytic devices (e.g., "El Teniente Converter" technology), the implementation of plants for the recovery of sulfuric acid and the management of gases from converters or furnaces.

The observed relationship between Chilean economic growth (Figure 5c), enforcement of national environmental regulations (Figure 5a) and trajectories for Hg, Pb, Cu, Mo, Sb, Nb and Sn (Figure 4), could lead to the wrong impression that socio-environmental history of the Puchuncaví valley fits into a "Environmental Kuznets

Curve" (EKC; Grossman & Krueger, 1993). This generalized hypothesis with evidence for and against (Arrow et al., 1995; Dasgupta et al., 2002) posits that during early stages of economic development the pollution rises, and then declines insofar the per capita income grows and environmental policies are adopted. Results presented here, however, suggest that reductions in seven metal(loid)s since 1998 CE cannot be simply viewed as environmental improvement since the aggregated pollution burden of long-lived pollutants (i.e., metal(loid)s) has increased substantially through time (Figure 5a). Moreover, the implementation of incremental actions through technology-based emission standards and decontamination plans appears insufficient for controlling the pollution load at VIA by persistent pollutants. This implies that environmental and health risks could have been locally amplified due to the synergistic and additive interaction of regulated short-lived pollutants (e.g., PM and SO_2) as well as long-lived pollutants that are not monitored or regulated by Chilean air quality policies. Such long-term pollution loads could lead to cumulative impacts that weaken the health and resilience of socio-ecological systems, and therefore, the environmental degradation may continue despite eliminating pollution sources (Arrow et al., 1995; Dasgupta et al., 2002). Thus, EKC lenses are unable to capture the complex -and non-linear- relationship between environmental quality and policing in the Puchuncaví valley. More importantly, this perspective masks key mechanisms that have molded the environmental injustice over this territory, including uneven redistributive policies, social exclusion, changes in zoning plans, national and international market forces.

5. Conclusions

Here, we highlight a cross-cutting approach capable of informing on how environmental injustice in sacrifice zones is intertwined with socio-economic and governance processes that operate at several scales -i.e., from local to transnational levels-. Actually, by conducting routine chemostratigraphic analyses, we afford a novel record for monitoring the magnitude, impacts and drivers (natural vs. anthropogenic) for sub-decadal variations in the accumulation of metal(loids) in the Puchuncaví valley over the past 136 years.

We demonstrate that the local socio-environmental history is best described by three pollution phases: 1872–1974, 1978–1997 and 2001–2008 CE. This area began to transform into a sacrifice zone at least from 1967 CE, and its evolution has been paced by the impressive growth in exports and economic wealth experienced by Chile since the late-twentieth century. Locally, this has resulted in an ever-growing pollution burden over the last 56 years brought by urban activities, but importantly the accelerated expansion and diversification of the industrial complex through increasing copper production and installed capacity of coal fired power generation. Although this staggering VIA development might have translated into an apparent reduction in local poverty levels in recent decades -i.e., reduced levels of deprivation-, such a path toward social equality is marginal. While such economic growth has been important for advancing the prosperity of the Chilean population, it has become counterproductive as it is maintained at the expense of the environmental degradation of the territory. Actually, the cost of pollution burden on the Puchuncaví valley has exacerbated throughout time affecting the community's well-being by reducing capacities to maintain traditional economies, a healthy life and to inhabit an environmentally secure territory. Hence, the pollution by metal(loid)s has become a patent Anthropocene risk that obstructs the process of local human development (sensu UNDP, 2021), which amplifies environmental injustice in the Puchuncaví sacrifice zone.

While national and international actions have apparently led to lagged and moderate lowering in some species, we verify that environmental curbing instruments have fallen short for reducing the pollution load in the area. The load of highly toxic metals (e.g., Pb, Hg, Cd, Cu) as well as of other trace metal(loid)s detected here after the implementation of control measures is far from approaching natural background levels reconstructed for the period prior to the development of VIA (1872–1960 CE). Environmental policies in Chile have operated under the preventive principle, so have emerged to decrease or moderate the impact that could result from exceeding emission standards for short-lived pollutants such as PM and SO_2 (Billi et al., 2021). In this sense, the comparison of metal(loid)s levels between the local pre-industrial period and the interlude 1960–2008 CE serves as a reference to inform on the amplitude for the change in pollution burden, and to envision more ambitious and targeted policymaking and actions. In fact, our results suggest that future governance structures aimed at managing the pollution hazards and socio-environmental conflict in the Puchuncaví sacrifice zone must not solely rely on the incremental introduction of less-polluting technologies, but also on the design of mitigation and adaptation strategies, including substantial citizen involvement. Specifically, focused on operational measures informed on ad hoc historical baselines -as provided by our PUCH0112-AT1 sediment core-for identifying feasible environmental rehabilitation potentials. This is in line with recommendations presented in a recent policy brief on socio-ecological governance for Chile (Billi et al., 2021), which stresses out the need of multiscale transformative and anticipatory actions to manage pollution drivers and impacts, and thus reduce vulnerabilities and increase resilience in territories.

Public and private actions embracing restoration are of utmost relevance for the Puchuncaví community -as other sacrifice zones-. Chile has recently updated commitments to Nationally Determined Contributions (NDCs; Gobierno de Chile, 2020) and recently submitted its Long-Term Climate Strategy (ECLP; Gobierno de Chile, 2021) to achieve just transition -i.e., the transition toward an environmentally and socially sustainable territories (Smith, 2017)-. Although both strategies propose concrete mitigation and adaptation actions -in terms of social equality and integration, emission of short-lived pollutants and carbon neutrality by 2050 CE- the just transition process for the Puchuncaví territory is not granted. The Ventanas1 coal-fired plant shut down in 2020 CE, and remaining VIA power plants are planned to be gradually decommissioned by 2035 CE. This decarbonization scenario might include or not an eventual closure of the Ventanas smelter-refinery. However, local communities will be still exposed to the pollution burden accumulated in the valley for the past 56 years, and most likely affected by the additive and synergistic effects of emergent long-lived contaminants and regulated short-lived pollutants. With the projected increase in extreme weather events in central Chile (Bozkurt et al., 2018), the management of the long-lasting pollution burden in the Puchuncaví valley is challenging. Increased frequency of floods, storms or droughts could lead to the spreading and remobilization of accumulated metal(loid)s (e.g., Guittonny-Philippe et al., 2014), thus bringing additional -or exacerbating- pollution issues either in local or nearby ecosystems and communities. Such latent hazard calls for urgent action and planning, as well as management measures that incorporate climate change adaptation and a just transition leading to sustainable and safe alternatives of income activities and sources. Otherwise, social and environmental inequalities in the Puchuncaví valley will continue to be perpetuated.

The feedback relationship documented here between environmental pollution, inequality, regulations and socio-economic trends have profound implications for understating how particular territories have been progressively and long lastingly impaired. This process appears in several regions around the world (Dionne, 2016; Lafratta et al., 2019; Li et al., 2018; Thevenon et al., 2011). Dong et al. (2021) stress out that there is not a consistent global upward trend in pollution by metal(loid)s through the twentieth century, but rapid increases brought by human activities have occurred localized around industrial areas. This reinforces the notion that the Anthropocene involves uneven burdens in specific territories that sustain production and consumption rates of a globalized world. Thus, our perspective for portraying the socio-environmental history in the Puchuncaví valley, comes to complement emergent initiatives that conceive sacrifice zones as spaces of the Anthropocene (Harrison, 2019; Tironi et al., 2018). In this vein, our work contributes to the discussion about how such localized Anthropocene risks persist and evolve mediated by the socially disparate, biased and increasing impacts of anthropogenic activities. We believe that our reconstruction based on a novel and high-resolution paleopollution archive represents a science-based instrument for informing the discussion on Anthropocene governance by providing data to evaluate the impact, design decontamination and environmental restoration plans, and extent of decision making on pollution control or to anticipate additional risks on polluted regions from climate change, and other hazards. More importantly, to advance in the assessment of environmental inequality in low-middle and upper-middle income countries, where development models still prioritize economic growth to the detriment of social and environmental security.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

Data sets with raw and processed data from elemental and geochemical analyses on the sediment core as well as socio-economic data are provided as Supporting Information (Data Sets S1–S4). All of these data sets are also available in the open-access repository Zenodo (https://doi.org/10.5281/zenodo.5805050; Gayo et al., 2021). Tables containing complementary results from statistical analyses (Tables S1 and S2) can be found in Supporting Information S1.

Acknowledgments

Funding for this work was provided by ANID/FONDAP/15110009, ANID-Millennium Science Initiative Program - NCN19_153, Grant ANID PIA/BASAL FB210006, Fondecyt projects #11150210, #11170566, #1201714, ESR UCV2095, PUCV DI 39.431/2020. We gratefully acknowledge the support of Isabel Cartajena and Diego Carabias in coring Los Maitenes wetland. The authors thank Yocelyn Santander for assisting sediment sampling, Francisca Flores for providing historical CASEN data for Puchuncaví and Michelle Ferrer for providing invaluable advice on the preparation of the plain language summary.

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