High resolution inventory of atmospheric emissions from transport, industrial, energy, mining and residential sectors of Chile

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Abstract. This study presents the first high-resolution national inventory of anthropogenic emissions for Chile (INEMA). Emissions for vehicular, industrial, energy, mining and residential sectors are estimated for the period 2015-2017 and spatially distributed onto a high-resolution grid (1 x 1 km approximately). The pollutants included are CO₂, NOx, SO₂, CO, VOCs, NH₃, and particulate matter (PM₁₀ and PM_{2.5}) for all sectors. CH₄ and Black Carbon are included for transport and residential sources, while Arsenic, Benzene, Mercury, Lead, Toluene, and Polychlorinated dibenzo-p-dioxins and Furan (PCDD / F) are estimated for energy, mining and industrial sources. New activity data and emissions factors are compiled to estimate emissions, which are subsequently spatially distributed using census data and Chile's road network information.

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- The estimated total annual national emissions of PM_{10} and $PM_{2.5}$ are 191 and 173 kilotonnes (Kt), respectively. The residential sector is responsible for over 90% of these emissions. This sector also emits 81% and 87% of total CO and VOC, respectively. Additionally, the energy and industry sectors contribute significantly to NH₃, SO₂, CO₂ emissions while the transport sector dominates NOx and CO₂ emissions, and the Mining sector dominates SO₂ emissions. In general, emissions of anthropogenic
- air pollutants and CO_2 in northern Chile are dominated by mining activities as well as thermoelectric power plants while in central the dominant sources are transport and residential emissions. The latter also mostly dominates emissions in southern Chile which has a much colder climate. Preliminary analysis revealed the dominant role of the emission factors in the final emission uncertainty. Nevertheless, uncertainty in activity also contributes, as suggested by the difference in CO_2 emissions between INEMA and EDGAR. A comparison between these two inventories also revealed considerable differences
- for all pollutants in terms of magnitude and sectoral contribution, especially for the residential sector. EDGAR presents larger emissions for most of the pollutants except for CH_4 and $PM_{2.5}$. The differences between both inventories can partly be explained by the use of different emission factors, in particular for the residential sector where emission factors incorporate information on firewood and local operation conditions. However, as mentioned above, differences in $C_{\text{pollutants}}$ emissions between both inventories also points to biases in the quantification of the activity.

This inventory (available at https://doi.org/10.5281/zenodo.4784286) (Alamos et al., 2021)) will support the design of policies that seek to mitigate climate change and improve air quality by providing policy makers, stakeholders and scientists with qualified scientific spatial explicit emission information.

1. Introduction

40 Air pollution is one of the main environmental challenges in Chile; in 2018 more than 9 million of its population (out of 17 million) were exposed to concentrations of fine particulate matter (PM_{2.5}) above the national air quality standard, and around 3,640 cases of premature mortality were estimated due to cardiopulmonary diseases (MMA, 2019a). Urban areas of central and southern Chile are among the most polluted in Latin America with important consequences for human health (Romero-Lankao et al., 2013) including an increase in hospital admissions and mortality associated with cardiovascular and respiratory 45 diseases (WHO, 2016).

The current air pollution and climate change problems are directly related to atmospheric emissions of criteria pollutants which affect air quality - and greenhouse gas (GHG). Identifying the origin and estimating the emissions of these pollutants by source type is a prerequisite for quantifying the impact of anthropogenic activity on air quality and climate, and thus

- 50 developing effective mitigation strategies. Additionally, having GHG emissions and criteria pollutants consistent with each other is key in the design of policies that allow addressing climate change and air quality in an integrated manner (Melamed et al., 2016).
- Currently, emission inventories of GHG in Chile are produced within the framework of their national determined contributions 55 (NDCs) as part of the commitments with the Parties to the United Nations Framework Convention on Climate Change (UNFCCC). Emission inventories of criteria pollutants are developed for the most polluted cities within the framework of the decontamination plans to develop mitigation strategies to improve urban air quality. The national GHG emissions are prepared by a team of professionals from the Ministry of Environment (MMA from Spanish Ministerio del Medio Ambiente) responsible for the development and update of the GHG emission inventories whereas the latter are prepared by consultants hired on a 60 case-by-case basis. Furthermore, while GHG inventories are performed consistently over the years, urban emission inventories of criteria pollutants are not necessarily consistent with previous versions and/or emission inventories of other cities. Additionally, the Register of Release and Transfer of Pollutants (RETC from Spanish Registro de Emisiones y Transferencia de Contaminantes) from the MMA gathers the emission declaration from the industrial sector and combines it with emission estimate from the residential and transport sector from different state agencies to build a national emission inventory. This 65
- information is available to the population through a dedicated web platform (www.retc.cl).

While the national GHG inventory provides annual emissions at a national and regional scale, inventories of criteria pollutants provide emissions at a conmunal level (RETC) or for an entire city (Decontamination Plans). However, these inventories (GHG and/or criteria pollutants) do not have the spatial nor the temporal resolution necessary for air quality modelling.

- 70 Regional air quality (AQ) assessments in South America have relied on global emission inventories to understand the interactions between emissions, air quality and public health (e.g. Longo et al., 2013; Rosario et al., 2013; Klimont et al., 2017; UNEP/CCAC, 2018). Furthermore, a comparison of global emission inventories against city-scale emission inventories for five South American cities (namely, Buenos Aires, Bogota, Lima, Rio de Janeiro and Santiago) revealed that although total emissions are in general comparable for these cities between the inventories, large differences exist for sectoral estimates
- 75 (Huneeus et al., 2020). Given that mitigation of air quality depends on identifying the dominating emission sectors, using global emission inventories is not recommended to define mitigation policies due to the risk of identifying the wrong target (Huneeus et al., 2020). Therefore, national inventories built on local data are needed to understand the contribution of human activity to air quality and climate change and design effective mitigation policies.
- 80 This paper presents the first gridded national inventory of anthropogenic emission for Chile of criteria pollutants as well as GHG (hereafter INEMA from Spanish Inventario Nacional de EMisiones Antropogénicas). The paper is structured as follows; the data and methodology used to estimate the emissions of each pollutant and sector are presented in section 2 while in section 3 the main results are shown, differentiating between the main pollutants and sectors that acquire relevance in the different regions of Chile. Discussion of the main results and uncertainty analysis of the estimated emissions are presented in section 4.
 85 Finally, in section 5 the main conclusions of this work are presented.

2. Methodology and data

The INEMA inventory includes yearly emissions of C bon Dioxides (CO₂), Nitrogen Oxides (NOx), Sulfur Dioxides (SO₂), Carbon Monoxide (CO), Volatile Organic Compounds (VOCs), Ammonia (NH₃) and Particulate Matter (PM₁₀ and PM_{2.5}) from the residential (section 2.2), industry (section 2.3), energy (section 2.3), mining (section 2.3) and transport sectors (section

90 2.4) for the years 2015 to 2017. Additionally, the residential and transport sector include emission estimates of Methane (CH₄) and Black Carbon (BC), while for the industry, mining and energy sector emissions of Arsenic, Benzene, Mercury, Lead, Toluene, and Polychlorinated dibenzo-p-dioxins and Furan (PCDD / F) are also reported. Emissions are grouped into sectors following the IPCC (2006) classification (Table 1).

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Sector considered on this paper	IPCC Code	IPCC Sector	Pollutants considered in our inventory
Energy	1A1	Energy Industries	CO, CO ₂ , VOC, NOx,
Industry	1A2 and 2, excluding 2C	Manufacturing Industries and Construction; Industrial Processes and Product Use	NH ₃ P 75, PM10, SO ₂ , Benzene, Arsenic, Toluene, PCDD/F, Mercury, and Lead
Mining	2C1, 2C2, 2C3 and 2C4	Metal Industry	
Residential	1A4b	Residential and others energy sector	CO, CO ₂ , NMVOC, NOx, NH ₃ , PM _{2.5} , PM ₁₀ , SO ₂ , CH ₄ and BC
Transport	1A3b	On road transport.	CO, CO ₂ , NMVOC, NOx, PM _{2.5} , PM ₁₀ CH ₄ and BC

Table 1 Pollutants and sectors considered in the Chilean inventory(INEMA) according to IPCC (2006) classification

100 The atmospheric emissions for each sector and pollutant are obtained by weighting the total fuel consumption (activity level) by an emission factor (EMEP / EEA, 2016), as shown in eq. (1).

$$E_{ijz} = \sum_{ijz} [NA_{ijz} * EF_{ij}] \tag{1}$$

where E_{izj} is the total emission for species or pollutant i on year z and sector j, NA_{ijz} is the activity level of pollutant i, in 105 sector j on year z, and *EF* is the emission factor for pollutant species i, type of source j. No interannual variability is assumed for the EFs. The following subsections present detailed methodology and considerations for each sector =

2.1 Study area.

Chile spans from 17 ° 29'57 "S to 56 ° 32'12" S and has a population of over 19 million inhabitants. The administrative political division is made up of 16 regions containing 56 provinces and 346 communes, presenting considerable differences in size and

110 population density. Furthermore, each commune contains urban and rural areas, with the exception of some purely urban communes in the larger cities. The territory can be broken down into 3 large macrozones with diverse climatic and geographical characteristics (Fig 1). The northern zone, with the regions of Arica and Parinacota, Tarapacá, Antofagasta, Atacama and Coquimbo, has an arid climate and includes the presence of the Atacama Desert, the driest desert outside polar regions (Rondanelli et al., 2015). Between 32- and 38-degrees south latitude is the central zone with the regions of Valparaíso, 115 Metropolitana, O'Higgins, Maule, Ñuble and Biobío. A Mediterranean climate with rainy winter and dry summer seasons prevails in this area. The regions of Araucanía, Los Ríos, Los Lagos, Aysén and Magallanes are located in the southern zone characterized by a temperate rainy climate, where low temperatures and abundant rainfall stand out. These conditions are accentuated further south, although the rainfall drastically decreases in the highest mountainous areas and south of the Strait of Magellan where a tundra-type climate predominates (Sarricolea et al., 2017).

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Figure 1 Continental Chile highlighting three macrozones defined for the paper, namely the northern (green), central (blue) and southern zone (red). Divisions within each macrozone indicate limits of the 1 ministrative regions.

125 2.2 Residential sector

Estimates from the residential sector include emissions produced by the combustion of biomass for heating, cooking and heating water. Firewood is acquired mostly through informal wood markets and the few regular and consistent information existing to characterize its consumption is collected through household surveys (REDPE, 2020). In this study, three studies (conducted in the last ten years) with regional representation are used to estimate the total firewood consumption in central

130 and southern Chile.

The first one of these three aforementioned studies was conducted by the Universidad Austral de Chile (UACH). Firewood consumption in the residential sector was estimated based on existing studies for the years between 2005 and 2012 for each region in southern and central Chile (UACH, 2013; hereafter UACH13). Another study was mandated by the Ministry of

- 135 Energy to the Corporation of Technological Development (CDT from Spanish Corporación de Desarrollo Tecnológico; https://www.cdt.cl), a private non-profit organization created in 1989 by the Chilean Chamber of construction. This study collected information on firewood consumption from the residential, commercial, public services, and industrial sectors for the entire Chilean territory for the year 2014. In each of the 16 Chilean regions, a total of 300 households in urban areas and 65 in rural ones were surveyed (CDT, 2015; hereafter CDT15). Finally, the most recent survey was done by the Forestry 140 Institute (INFOR from Spanish Instituto Forestal) collecting samples for between 300 and 500 households for a single year between 2015 and 2018 in the 6 Central-Chilean regions considered in the study (INFOR, 2019; hereafter INFOR19) (Fig 2).
- Despite their differences, these studies agree that the consumption increases towards the south consistent with lower temperatures and the corresponding higher energy requirement of dwellings (Fig 2). For several individual regions, however, significant differences in wood consumption are four \overline{z}

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Activity levels in INFOR19 are higher than the ones estimated in CDT15 and methodological shortcomings that potentially explain this underestimation have been identified (Reyes et al., 2018; Reyes 2017). For this reason, firewood consumption from CDT15 is only used for regions lacking alternative data (all regions north of Santiago). For the regions of O'Higgins, Maule, Bio-Bio, Ñuble, Araucanía and Los Ríos, the data reported in INFOR19 are used, whereas for regions Los Lagos, Aysén, and Magallanes, the information from UACH13 is selected over CDT15. Consumption estimates from UACH13 are consistent with the results from INFOR19 for regions with data from both sources (Fig 2). We highlight that only INFOR19 provides firewood consumption at the communal level, the remaining studies estimate the firewood consumption (AHFC) is computed at a communal level. For regions where the data are available at a regional level, the same average consumption is

assumed for all communes contained in the administrative region.

A bottom-up approach is used to standardize the different information sources for the study period. The activity level of residential emissions is obtained at communal level (c), differentiating between urban and rural areas (a) as follows:

$$A_{y,c,a} = HN_{y,c,a} * PF_{y,c,a} * AHFC_{c,a}$$
(2)

160 Where $A_{y,c,a}$ is the residential fuelwood consumption for the year y in commune c for urban/rural area a; $HN_{y,c,a}$ is the number of total dwellings in year y in commune c in area a registered in INE (2019); $PF_{y,c,a}$ is the "penetration factor" representing the percentage of houses that use biomass in the year y in commune c and area a obtained from the Chilean Ministry of Social Development (MDS from Spanish Ministerio de Desarrollo Social; 2015; 2017) and $AHFC_{c,a}$ is the average household firewood consumption in commune c and area a. The MDS conducts every two years the National Socio-Economic Characterization Survey (CASEN) for the entire country. This survey contains information on the type of fuel used by households for heating, cooking food, and domestic hot water production, allowing to derive the penetration factor (PF) of biomass. For those isolated communes where this survey is not applied, the PF is taken for each region at urban or rural level considering the regional PF value from CDT (2015).



Figure 2 Total fuelwood consumption on kilotonnes by region according to UACH (2013, red), CDT (2015, green) and INFOR (2019, blue). Regions are colored according to the data source used in each region.

- 175 In this study, we use the local EFs from SICAM (2014) and also used in MMA (2019b) to estimate emissions of NOx, PM_{2.5}, PM₁₀, and SO₂ (Table A1 in Supplementary Material) and those estimated from MMA (2019b) for CO₂ and NH₃. Among other factors, the EFs vary according to the efficiency of the technology used (e.g., fireplace, wood stove, simple heater, heater with temper, etc.), the humidity present in the wood, and the device's operating conditions¹ (Jimenez et al., 2017; Guerrero et al., 2019; Schuefftan et al., 2016). For CO and NMVOC's (Non-Methane VOCs) we use EFs for dry firewood from EMEP/EEA
- 180 (2019;) while for CH₄ the EFs estimated on the tier 1 approach from IPCC (2006) are used. However, EMEP/EEA and IPCC's database do not present EFs for wet firewood and bad operation of heating devices. To compensate for this, we follow USACH (2014) and apply correction factors of 1.5 for wet firewood combustion and 3 for poor operation. Finally, we follow EMEP/EEA (2016) and consider BC to represent 10% of PM_{2.5} emissions.

2.3 Transport sector:

- 185 Estimated emissions from the transport sector consider exhaust emissions from vehicles traveling on public routes nationwide, in urban and interurban areas, for years 2015 to 2017. Rail, air and sea modes are not included, nor are off-road machinery. Emissions were calculated per region based on estimates of number of vehicles and their activity level. A more detailed description of the method applied to estimate transport emissions can be found in Osses et al. (2021, this issue)
- 190 The different types of vehicles and their activity levels per region come from information obtained from official reports of government agencies. This information includes statistics on fleet composition as the number of registered vehicles by region (INE 2017b), average annual mileage by vehicle type (SCSS, 2014; MAPS, 2013) and fuel sales for road transport by region (SEC, 2017). Vehicle categories considered are: light passenger, commercial and taxi vehicles; 12- and 18-meter buses; light, medium and heavy duty trucks; and two-wheeled vehicles. Each of these categories is subdivided according to the type of fuel used (gasoline or diesel) and the emission standard in its European equivalent (EURO standard). Total fuel consumption of registered vehicles was estimated and compared to real fuel sales for each region. Thus, the number of active vehicles in a region was inferred and the number of vehicles per region was adjusted accordingly. The distribution of vehicles into urban and interurban activity per region was based on a proportional regional distribution provided by SCSS (2010). The combination of categories, fuels and emission standards generates a total of 70 types of vehicles for the emission analysis, distributed 200 regionally and distinguishing between urban and interurban activity.

¹ A bad operation condition occurs when combustion is carried out with the stove draft closed.

² "Any copper smelter or any other industrial source emitting ash where a thermal treatment of mineral or metallurgical compounds of copper and gold is carried out, whose ash content in the feed is greater than 0.005% by weight on a monthly basis" taken from the Decree 28 on Emission Standard from copper smelters and arsenic emission sources.

Activity level was expressed in *VKT* (vehicle kilometre travelled) calculated as the sum of the number of vehicles per mileage per type of vehicle (eq. 3) expressed as follows

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$$VKT = \sum_{i,j,k}^{N} * N_{i,j,k} * KM_{i,j,k}$$
(3)

where $N_{i,j,k}$ is the number of vehicles of type i in region j and road class k (urban or interurban); $KM_{i,j,k}$ are the kilometers travelled per year by vehicles type i, in region j and road class k.

The estimate considers that all vehicles that enter Chile are required to comply with the European EURO regulations or their 210 US equivalent. Consequently, the assignment of emission factors for each of the vehicle types was carried out by applying COPERT V values (EMEP/EEA, 2019), adapted to the Chilean fleet (Gomez, 2020). Total emissions are calculated by multiplying *VKT* by an emission factor in grams per kilometer. The result is a regional emission database distinguishing by urban and interurban emissions, for CO, CO₂, VOC, NOx, PM_{2.5}, PM₁₀ CH₄ and BC.

2.4 Point sources: Energy, Mining and Industry sectors

- Emissions from point sources and for species listed in table 1 are not estimated by our work but downloaded from the Register of Emissions and Pollutant Transport (RETC from spanish Registro de Emisiones y Transporte de Contaminantes, <u>https://datosretc.mma.gob.cl/group/emisiones-al-aire</u>). from the Ministry of Environment. This register receives all self-reported emissions by the industrial facilities in accordance with current environmental regulations (MMA, 2019b). Industries are not obliged to declare CH₄ and NMVOC but only total VOCs. Therefore, when analysing VOC emissions from the energy, mining and/or industry sector we will be referring to total VOCs.
 - The facilities that must declare their emissions ar
 - Pulp and Paper Production, Primary and Secondary Smelters, Thermoelectric Power Plants, Cement, Lime and Gypsum Production, Glass Production, Ceramic Production, Iron and Steel Industry, Petrochemical Industry, Asphalt Production.
 - Industries with generator sets greater than 20kW, and industrial and heating boilers with fuel energy consumption greater than 1 Mega Joule per hour.
 - Establishments with electricity generation units, made up of boilers or turbines, with a thermal power greater than or equal to 50 MWt
 - Establishments whose fixed sources, made up of boilers or turbines, individually or as a whole, add a thermal power greater than or equal to 50 MWt
 - Establishments corresponding to copper smelters and arsenic emitting sources

In this work, emissions from point sources are differentiated between Energy, Mining and Industry sectors. The energy sector includes production and distribution of fuels and the generation of electric energy while Mining includes the production and

235 smelting of metals. The remaining point sources will be aggregated into a single sector to which we will refer as Industry henceforth.

This database includes more than 8324 point sources along the territory, most of which have associated coordinates. Those without a specified location were pinpointed on google earth using the facilities names and the address provided in their declaration if their contribution to the total communal emission was larger than 20% point sources without a geographic

240 location contributing with emissions less than the above threshold were not explicitly included in the inventory, however their emissions were distributed among the located sources. For a given species and sector and within each commune, emissions of located sources were scaled to fit total (located + non-located) emissions in that commune and sector.

2.5 Spatial distribution of emissions:

While point source emissions from Industry, Mining and Energy sector are spatially distributed using their coordinates (section
2.4), those from the transport and residential sectors are estimated at the regional or communal level and thus need to be distributed to the final distribution of 0.01°x0.01° (approximately 1x1 km) by means of a proxy (Fig 3).

Residential emissions were initially estimated at the communal level and distributed onto a regular 0.01°x0.01° grid (approximately 1x1 km) based on pop test on density from the last census conducted in 2017 (INE 2017a) available at census block scale. The latter is the smallest territorial scale for which relevant information from the census exists, it consists of a group of adjoining or separate dwellings, buildings, establishments and/or properties, delimited by geographical, cultural and natural features. The population distribution was obtained by projecting the 0.01°x0.01° lat-lon grid (epsg:4326, WGS 84) onto

The spatial distribution of transport emissions within each region was performed by projecting the road network of each region onto the grid with cells of 0.01x0.01 degrees of latitude and longitude (approximately 1x1 km). QGIS open source software, the official database for Chile's road network and regional limits (BCN, 2020) was used to characterize the Chilean road network and was complemented with information from Open Street Map (OSM, 2020) to organize it into a hierarchy comprising freeways, arterials, collectors, and local roads of 77800 km of both rural lands and main citi standard vehicle flow

the information contained in the census blocks and retrieving the aggregated information.

260 per type of road was estimated applying a road weight factor, based on toll barrier vehicle counts at interurban roads and origin-destiny surveys in urban roads. Average weight factors are 54% for freeways, 23% on arterials, 16% for collectors and 7% on local roads. In each region, urban emissions were distributed among cities based on population (INE, 2017a) first and then within each city emissions were distributed applying the aforementioned weight factors. Thus, urban emissions in each cell depend on the population and the roads in the cell.

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Figure 3 Map of Santiago at different scales: in blue the utilized $0.01^{\circ} \ge 0.01^{\circ}$ grid (approximately 1x1 km), and in black the $0.1^{\circ} \ge 0.1^{\circ} \ge 0.1^{\circ}$

3. Results:

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Total national emissions increase for PM_{2.5}, NH₃, S and CO₂ between 2015 and 2017, whereas CO, VOC, PM₁₀ and NOx decrease during that period (Fig 4). While PM₁₀ decreases due to decreasing trend in industrial emissions and stable residential ones, NOx remains mostly constant due to a decrease in energy emissions and the slight increase in the rest of the sectors. Almost half of the NOx emissions are from the transport sector while the industry and energy sector combined contribute to almost an equivalent amount of the total NOx emissions. Although the largest contributor to CO₂ emissions in Chile is the Energy sector due to thermoelectric power plants (MMA, 2017), the increase is associated with the increase of CO₂ emissions in Chile. This activity drives the increase in SO₂ emissions from 2015 to 2016. However, the decrease from 2016 to 2017 is the result of reductions in the energy and industry sector combined with mostly stable emissions from the mining activity. Finally, for NH₃ the emissions are dominated by the energy and industry sectors. We highlight that the agriculture or is not

included in this inventory and therefore not reflected in emissions of CH_4 nor NH_3 . In general, the agriculture sector dominates NH_3 and CH_4 emissions and for any future study on these species, these sectors would need to be included.



Figure 4 Total national annual emissions distributed by sector for pollutants VOC, PM_{2.5}, PM₁₀, NOx, NH₃, CO₂, CO, and SO₂ in kilotonnes for 2015-201

Emissions of NOx dominate in Central C_{12}^{12} due to larger urban centers and vehicular traffic in this area while CO₂ emissions are distributed mainly in north and central Chile, where thermoelectric power plants are abundant (Table 2). Furthermore, PM_{2.5} and PM₁₀ are mostly emitted in southern Chile with large contributions also in central Chile, mainly from the residential

290 sector in both cases. This sector also makes the largest contribution in CO and VOC. SO₂ has a greater presence in the northern part of the country, consistent with a larger mining activity.

Pollutant	Tot:	North	Centre	South
BC	15.6	3.2%	35.3%	61.5%
СО	1477	2.4%	39.5%	58.1%
CO_2	85402	29.6%	52.6%	17.9%
NH ₃	23	26.4%	45.7%	27.9%
NOx	2132	26.4%	49.9%	23.7%
PM ₁₀	191	4.6%	35.2%	60.2%
PM _{2.5}	173	3.1%	35.3%	61.6%
SO_2	261	62.3%	35.4%	2.2%
VOC	256	2.2%	35.7%	62.1%

Table 2 Average total emissions from Energy, Industry, Residential, and Transportation sectors [in kilotones] in the 2015-2017 period by pollutant and macrozone.

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Given the large health impact associated toPM2.5 and its role in poor air quality in central and southern Chile, we focus now on this particular pollutant and its spatial emission distribution along the territory (Fig 5). More than 90% of the 158 (170)
Tors of PM_{2.5} (PM₁₀) total national emissions for 2017 originated from the residential sector (Fig 5). Emissions in the northern macrozone are mostly from the energy and industry sectors, which are generally located in urban areas. The Mejillones
commune concentrates more than 20% of all PM_{2.5} emissions in the northern macrozone (Fig 6a). More than 1,300 tons are emitted per year in this commune, of which 99% come from the energy sector (thermal power plants) concentrated in a few locations.

In Central and Southern Chile emissions are largely dominated by the residential sector and are consequently distributed along the territory according to population, with a larger magnitude in locations with a greater number of dwellings and concentrated in the country's central valley. However, contrary to cities of Southern Chile (Fig 6c), significant contributions from other sources are observed in some areas of Central Chile. For instance, Santiago, the capital of Chile (Fig 6b), where more than 40% of the country's population resides, stands out in Central Chile. Although firewood burning for heating and cooking is prohibited in the metropolitan area, it is still the largest contributor to PM_{2.5} in the region due to its use in the outskirts and surroundings; from the 2030 H PM_{2.5} emitted, 1480 Kt are emitted in the outskirts and surroundings of the city. Within Santiago, the largest polluter is the transport sector, representing 22% of the total $PM_{2.5}$ emissions and almost 90% of the 3 annual Kt of NOx.



Figure 5 Spatial distribution of the 2017 emissions of particulate matter of the Energy (red), Industry (green), Residential (blue) and Transportation (purple) sectors in a grid of 100 ° x 0.01 ° on a map of Chile according to the macrozones defined for the country (Figure 1). Note that the Metropolitan region includes the capital Santiago and ~ 40% of the population. The pie charts indicate the relative contribution that each source makes to total PM_{2.5} emissions in each region.



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Figure 6 Spatial distribution of the 2017 emissions of $PM_{2.5}$ of the Energy (red), Industry (green), Residential (blue) and Transportation (purple) sectors in a grid of 0.01 x 0.01 degrees (approximately 1x1 km) for Mejillon $\overline{F}(4)$, Santiago (b) and Temuco and Padre Las Casas Cities (c). These cities are located in the Northern, Central and Southern zones, respectively.

325 3.1 Comparison of total emissions by sector and pollutant with EDGAR inventory

Puliafito et al. (2017) and Huneeus et al. (2020) show that despite consistencies in the magnitude of total emissions of pollutants, global inventories have large discrepancies in sectoral contribution when compared to local or national inventories. We compare estimated emission for 2015 from the present inventory against the EDGAR v5.0 in ory (Crippa et al., 2019; 2020). Global inventories, such as EDGAR, have been used in South America in the absence of a local inventory for AQ assessments (Huneeus et al., 2020). Both inventories, EDGAR and this work, follow the sectoral classification proposed in IPCC (2006) allowing thus the direct comparison of sectoral emissions.

The differences for 2015 between both inventories for all pollutants are considerable in terms of magnitude and sectoral contribution, especially for the residential sector (Fig 7 and 8). Except for PM_{2.5} and CH₄, EDGAR presents larger emissions than INEMA. For CO, VOC and CO₂, emissions are between 20 and 40% larger, for NOx differences are around 90%, while for PM_{2.5} INEMA emissions are 45% larger than those estimated on EDGAR. These differences can partly be explained due to the use of different emission factors in both inventories. The election of emission factors for residential wood burning which include filterable PM only or filterable PM + condensable PM have an extremely large influence on the estimated emissions (e.g. Denier van der Gon et al.,2015). Moreover, the emission factors do not consider wet firewood and combustion with poor operating conditions which considerably increases these pollutants' emissions (Schueftan et al., 2016; Guerrero et

al., 2019). The current inventory (INEMA) however, considers emission factors that take these conditions into account. For NOx the larger emissions in EDGAR are from the transport, energy and industry sector. While EDGAR and INEMA have comparable CO_2 emissions for the transport, mining and residential sector, they differ significantly for the Energy and Industry sector. We note that the estimated emissions in this work for the energy and industry sectors are in line with what Chile reports

345 to the UNFCCC (34*10³ KT for energy and 18*10³ for industry) (MMA, 2020) suggesting a potential source of bias in the activity data used in EDGAR for the energy sector.

EDGAR VOC transport emissions are larger, due to evaporation emissions, which are not considered on INEMA inventory. Furthermore, smaller EDGAR emissions of PM_{2.5} and VOC are mostly due to differences in emissions from the residential sector. Although the use of distinct EF by both inventories might explain this discrepancy, differences in estimating activity as highlighted by different CO₂ emissions might also explain part of the difference.

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Figure 7 Emistric s of CH₄, CO, CO₂, NMVOC, VOC, NOx, PM_{2.5} for 2015 reported by EDGAR v5.0 (purple) relative to the present work (INEMA; pink). EDGAR emissions were normalized by the magnitude of the current INEMA inventory.



Figure 8 Total 2015 emissions in kilotonnes by pollutant and sectors according to this work and EDGAR v5.0. Sectors considered in both inventories correspond to the classification proposed in IPCC (2006) presented in section 2.

360 4. Uncertainty and quality of estimations on the residential sector

Emissions represent a large source of uncertainty in air quality modelling (Thunis et al., 2016) of which uncertainty in emission factors dominate over the better-known activity data (Scarpelli et al. 2019). Consistently, for the residential sector in this study, the largest uncertainty in the estimated magnitude is associated with the emission factor. For VOC, CO, BC and particulate matter emissions, the range of possible estimations using varying information sources can reach differences of a factor 84, 24,

- 365 13 and 13 respectively (Fig 9a). For VOC and PM not all differences can be attributed to uncertainty, it is partly related to the choice of what is included in the definition of VOC or PM. In the case of PM_{2.5} emissions, differences in the estimated magnitude following the different emissions factors can reach up to factor 8 whereas differences in the activity data are less than a factor two (Fig 9b). However, the final uncertainty is even larger when considering the combined uncertainties from each parameter (Fig 9b). CO₂, NH₃, and SO₂, have lower uncertainty ranges (Fig 9a) due to the greater consensus on their EFs
- 370 in the literature (MMA,2019b; IPCC, 2006; US EPA (1996a; b); SICAM, 2014) and the fact that these are single well-defined species whereas VOC and PM are container definitions; they include a variation of species or sizes. For CO₂, NH₃, and SO₂ the possible estimations of the lower and upper limits differ by a factor 2, while for NOx this value can reach up to a factor 5.



Figure 9 a) uncer and y range of residential emissions considering the possible estimation that can be made with the different sources of information evaluated in this work. b) PM_{2.5} uncertainty range disaggregated according to different emission factors (groups of columns) and activity levels (colors). In yellow are the inventories constructed using CDT information (CDT, 2015), which provides the lowest possible activity level (AL) while in red are the activity levels used in this inventory (section 2.1). The first group of columns represent estimated PM_{2.5} emissions based on EF from RETC while the second group correspond to the estimate based on EF used in the current inventory. The third and fourth group of columns correspond to the estimate based on EF proposed by US EPA (1996a;b) and EMEP/EEA 2019, respectively.

The final EF's used in this study results from aggregating several EF each one of which corresponds to specific emission conditions, and/or fuel components that determine the magnitude of the emitted flux, by weighting each EF according to distribution parameters estimated on household surveys. The more levant parameters considered are, among others, the quality and efficiency of the used technology (appliance type), the tree species, the humidity of firewood fuel, and the operating conditions of devices (Jimenez et al., 2017; Guerrero et al., 2019; Schuefftan et al., 2016). Each of these EFs has its uncertainty, which depends on the quality and the number of laboratory tests carried out to determine its robustness (RTI International, 2007). Despite the magnitude of data and studies carried out, the uncertainty associated with EF estimation is considerable. EMEP/EEA 2019 indicates that for a standard heater, the associated uncertainty to the estimated CO and PM_{2.5} EF can be

larger than ten and four times, respectively. Additionally, estimating activity data has also sources of uncertainties associated.

390 For instance, to estimate the amount and type of firewood consumed as well as the technology used and operating conditions, household surveys are usually conducted. These studies have big uncertainties, due to high informality surrounding the firewood transactions and markets (Zhao et al., 2011), and depend on the size of the surveyed sample as well as its representativity. The combination of the aforementioned uncertainties (Fig 10) impacts the expected final confidence of aggregated EFs and thus the magnitude of the estimated emissions (EMEP / EEA, 2016)



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Figure 10 Components that determine the final uncertainty of residential emission estimations

5. Data availability

The emission database described is available at Zenodo (https://doi.org/10.5281/zenodo.4784286) (Alamos et al., 2021). The database consists of one .tar file for each year and sector, containing NetCDF (.nc) files for each pollutant. Each NetCDF contains annual total emissions for the pollutant and year indicated per grid cell

6. Conclusions

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A high-resolution emission inventory $(0.01^{\circ} \times 0.01^{\circ} \text{ degrees}, \text{ approximately } 1 \times 1 \text{ km})$ of criteria pollutants, CH₄ and CO₂ from transport, industry, mining, energy, and residential sectors in Chile for the period 2015 to 2017 was developed. This is the first time a national gridded emission inventory with consistent CH₄, CO₂ and criteria pollutants was created for the entire

- 405 country. Urban and rural emissions from the residential sector are estimated based on firewood consumption data derived from different surveys conducted at the regional and communal level. The transport sector includes vehicles travelling on public urban and interurban routes nationwide. For mining, industry and energy sources, the self-reported emission estimates compiled by the environmental agency RETC are used.
- 410 While Total national emissions increased for PM_{2.5}, CO₂, NH₃ and SO₂ between 2015 and 2017, they decreased for CO, VOC, PM₁₀ and NOx. Estimated total annual emissions for the period 2015-2017 for PM₁₀ and PM_{2.5} were 192 and 173 Kt, respectively, of which more than 90% is emitted by the residential sector. This sector is also responsible for 98% of the 1923 Kt CO emissions and 88% of the 710 Kt VOC emissions. Regarding NOx, total average annual emissions were estimated at 213 Kt and dominated by the transport sector (87 Kt) while the industry and energy sector combined contribute an almost equivalent amount to the total NOx emissions with 57 and 36 Kt, respectively. Additionally, CO₂ emissions (85,402 Kt) are
- dominated by the Energy sector mostly due to emissions from thermoelectric power plants (33,911 Kt) followed by the transport and industrial sectors (22,770 and 13,804 Kt, respectively). Mining activity (due to copper smelters) dominates SO₂ emissions in Chile, contributing an average of 201 Kt of a total of 294 Kt in 2017.
- 420 A comparison of the estimated emissions against the EDGARv5 database (Crippa et al., 2019; 2020) shows significant differences for several species. For CO, VOC and CO₂, EDGAR emissions are between 20 and 40% larger, while NOx have differences around 90%. On the other hand, PM_{2.5} emissions estimated in this work are 45% larger than those estimated on EDGAR. These differences are even larger when considering emissions per sector, in particular for the residential and energy sector. Furthermore, a preliminary uncertainty analysis on the residential sector suggests that the main uncertainty source in
- 425 this work is the diversity in emission factors available, calling for the need to compile emission factors that include local conditions. Uncertainties also exist in activity data as suggested by the difference in CO_2 emissions between INEMA and EDGAR.

The dominant contribution of the residential sector to various pollutants, especially particulate matter, highlights the importance of increasing efforts to mitigate this source. Increasing the thermal efficiency of dwellings, improving the firewood combustion quality by reducing the humidity of the burned wood or increasing the efficiency of combustion technologies, and implementing educational campaigns that ensure the correct use of the devices, are among the potential policies to achieve this goal. Nevertheless, a consistent and robust estimation of firewood consumption is prerequisite to estimate emissions from the residential sector. This requires the creation of an official database that characterizes firewood consumption throughout the territory. Given the timeliness of the consumption data used in the present work, the absence of such an official database would

prevent updating the present inventory in the near fut

This is the first version of a national gridded inventory that will need to be further developed and continuously updated. It can be an important reference and benchmark for comparison in the future to track the impact of mitigation or other policy

440 measures. Further, future development of this inventory should consider, for instance including the agriculture sector and offroad vehicle emissions as well as completing the industry sector by locating in the territory the non-documented sources. Nevertheless, this inventory provides policy makers, stakeholders and scientists with qualified scientific spatial explicit emission information to support air quality modelling and the development and further evaluation of policies to minimize (health and climate relevant) atmospheric pollutant emissions.

445 Author Contributions

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NA and NH lead and write the original draft with contributions of all authors. NA, NH, MOpazo, SP prepare and curate the data, MOsses and NP generate and describe the emissions from transport sector. RR and AS participate in the processing of the residential activity level data. NA generates the residential emission data while NA and MOpazo prepared the industry, mining and energy emission estimate. HDvdG and NH designed the algorithm to distribute the residential emissions and together with RC provide feedback on the methodology used and the global consistency of the inventory. All the authors review and edit the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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Appendice

Table A1 shows emission factors for the residential sector by technology (appliance type), humidity and operation device conditions for PM₁₀, PM_{2.5}, NOx, and SO₂. For CO and NMVOC's (Non-Methane VOCs) we use EFs for dry firewood from EMEP/EEA(2019;) while for CH4 the EFs estimated on the tier 1 approach from IPCC(2006) are used.

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Table A2 shows the aggregated emission factors (g/kg) at a regional level for each pollutant used in the residential sector (eq A1), considering the distribution of technologies and humidity conditions of fuelwood estimated in CDT (2015) for each region, and 30% of devices bad operation, according to RETC (MMA, 2019). For CO_2 and NH_3 are used EFs estimated from MMA (2019b)

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$$EF_{ij} = \sum_{z} \sum_{x,j} \sum_{y,j} \left[\left[\left[EF_{i,x,y,z} * T_{y,j} \right] * H_{x,j} \right] * OD_z \right]$$
(A1)

Where:

 EF_{ijz} is the emission factor for pollutant *i* on region *j*

 $EF_{i,x,y,z}$: Is lab emission factor estimated for pollutant *i*, humidity conditions of fuelwood *x*, technology for combustion *y* and quality operation device *z*

660 $H_{x,j}$: is the proportion of humidity conditions x of fuelwood on region j

 $T_{y,j}$: is the proportion of technology conditions for fuelwood combustion y on region j

 OD_z : is the proportion of bad operation device condition z considered at a national level

Technology	Pollutant	Dry	Wet	Bad operation

Cook stoves	CH_4	4,5	7,11	13,5
	NMVOC	9	14,22	27
	PM_{10}	7,5	13,9	33,8
	PM _{2.5}	7,0	12,9	31,5
	СО	60	94,8	180
	NOx	2,1	2,7	2,7
	SO_2	0,2	0,2	0,2
Conventional	CH ₄	4,5	7,11	13,5
stove	NMVOC	9,0	14,22	27
	PM_{10}	6,2	11,8	45,8
	PM _{2.5}	5,8	11,0	42,6
	СО	60	94,8	180
	NOx	2,0	3	3
	SO_2	0,1	0,2	0,2
Catalytic stove	CH ₄	4,5	7,11	13,5
	NMVOC	5,7	8,927	16,95
	PM_{10}	5,2	11	29,5
	PM _{2.5}	4,8	10,2	27,5
	СО	60	94,8	180
	NOx	1,9	2	2
	SO_2	0,1	0,1	0,1
Open fireplace /	CH_4	4,5	4,5	-
Others	NMVOC	9,0	9,0	-
	PM_{10}	12,7	28,5	-
	PM _{2.5}	11,8	26,5	-
	СО	60	94,8	_
	NOx	7,7	3,1	-
	SO ₂	0,2	0,2	-

Table A1 emission factors for the residential sector by technology (appliance type), humidity and operation device conditions

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Region	PM _{2.5}	PM ₁₀	SO ₂	CO	NOx	VOC	CO ₂	NH ₃
North Zone	14,45	15,54	0,15	134,60	3,78	72,72	1366,83	0,87
Valparaíso	14,05	15,03	0,13	123,71	3,21	59,05	1366,83	0,87

O'higgins	15,60	16,76	0,16	149,70	3,74	83,95	1366,83	0,87
Maule	13,80	14,82	0,14	128,33	3,13	58,27	1366,83	0,87
Bio-Bio	15,07	16,19	0,15	144,12	3,10	68,15	1366,83	0,87
Araucania	17,80	19,12	0,14	178,02	2,44	75,03	1366,83	0,87
Los Ríos	15,86	17,04	0,15	177,95	2,50	67,02	1366,83	0,87
Los Lagos	15,68	16,84	0,14	173,604	2,39	61,02	1366,83	0,87
Aysén	15,74	16,91	0,15	185,81	2,38	64,51	1366,83	0,87
Magallanes	17,32	18,60	0,15	171,62	2,77	77,63	1366,83	0,87
Metropolitana	16,79	18,04	0,14	150,16	2,76	71,68	1366,83	0,87
Mean	15,65	16,81	0,15	156,15	2,93	69,00	1366,83	0,87

Table A2 aggregated emission factors (g/kg) at a regional level for each pollutant used in the residential sector.