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Abstract

The changing composition of the atmosphere, driven by anthropogenic emissions, is the cause of anthropogenic climate change as well as deteriorating air quality. Emission inventories are essential to understand the contribution of various human activities, model and predict the changing atmospheric composition, and design cost-effective mitigation measures. At present, national emission inventories in South America (SA) focus on Greenhouse Gases (GHG) as part of their obligations to the United Nations Framework Convention for Climate Change (UNFCC) within the framework of their national communications. Emission inventories other than for GHG in SA focus mainly on growing urban areas and megacities. Therefore, studies examining air quality at national, regional or continental scales in SA depend on (down-scaled) global emission inventories. This paper examines the emission estimates of air pollutants from various global inventories for five SA countries, namely Argentina, Brazil, Chile, Colombia and Peru. A more detailed analysis is conducted for the EDGAR and ECLIPSE emission inventories, in particular against local city-scale inventories of a major city in each country. Although total emissions between down-scaled global inventories and local city inventories are often comparable, large discrepancies exist between the sectoral contributions. This is critical as the mitigation of poor air quality will depend on addressing the right sources. Potential sources of discrepancies between global and local inventories include the spatial distribution proxies, difference in emission factors used and/or the use of generic statistical country data when estimating emissions. This highlights the importance of using local information when generating national emission inventories, especially for air quality modeling and development of effective mitigation measures. This work represents a first step towards an increased understanding of the strength and weaknesses of emissions information in SA.

Key words;

South America, anthropogenic emissions, air pollutants, Argentina, Brazil, Chile, Colombia, Peru, city emission inventories

1 Introduction and Rationale

Over the last decades, environmental problems such as acidification, eutrophication, air pollution and climate change have caused significant adverse impacts on the environment, human health and vegetation (Steffen et al, 2015; HTAP, 2010; Schneidemesser et al., 2015). These environmental problems are directly related to the atmospheric emissions of greenhouse gases (GHG), air pollutants and their precursors. Reliable emission inventories are a prerequisite to understand these environmental issues including the impact of anthropogenic activity on air quality and climate, and to develop effective mitigation options. Furthermore, emission knowledge of GHG and air pollutants are key in the development of integrated policies addressing climate change and/or air quality (AQ) and reducing unintended consequences (Melamed et al., 2016; Schmale et al., 2014; Reis et al., 2012).

Emission inventories developed in different South American (SA) countries at national level typically focus on GHGs as part of the obligations of the Parties to the United Nations Framework Convention on Climate Change (UNFCCC) within the framework of their national communications (Baumgardner et al., 2018). Emission inventories other than for GHG in SA focus mainly on megacities in an effort to understand the interactions and feedback mechanisms between emissions, AQ and public health (Alonso et al., 2010; Gallardo et al., 2012a). Except for Argentina (Castesana et al., 2018; Puliafito et. Al., 2015; 2017), no national emission inventory for air pollutants is available with the spatial and temporal detail needed for AQ modelling, analysis and policy support. Therefore, the emission inventories currently used for national, regional or continental scale AQ assessments in SA are derived from global data sets (e.g. Longo et al., 2013; Rosario et al., 2013; Kumar et al., 2016; UNEP/CCAC, 2018).

South America is a continent that spans over the northern and the southern hemisphere, from the very cold Tierra del Fuego, close to Antarctica, to the equator and beyond to the Caribbean Sea. Its climate exhibits tropical, subtropical as well as extratropical features (Garreaud et al., 2009). It includes the world's largest rainforest, what is considered the driest desert outside polar regions (Rondanelli et al. 2015) and the Andes mountain range peaking well above 6000 meters, introducing east-west climate asymmetries (Garreaud et al., 2009). In short, a very diverse collections of ecosystems, physical landscapes and climate zones. While SA countries experienced similar population growth in the last 20 years, there have been large differences in economic development. Emissions sources of air pollutants dominating impact on air quality also vary largely from country to country from residential combustion in central and southern Chile (Saide et al., 2016; Mazzeo et al., 2018) to transport in Colombia (Gonzalez et al., 2017; Pachon et al., 2018). Furthermore, emission conditions also vary largely, in particular among main metropolitan areas; while pollutants are emitted at sea level in Buenos Aires (Argentina), those in La Paz (Bolivia) are emitted at approximately 3600 m above sea level. This altitude can have an important impact on vehicle emissions with different emissions due to altitude (He et al., 2011; Ni et al., 2014; Szedlmayer and Kweon, 2016; Wang et al., 2018).

The available global emission inventories for selected countries are analyzed by source-sector in this paper. These countries are highly urbanized, except for Peru, urbanization is above 80% and more than 50% of the urban population of these countries live in cities larger than 300.000 inhabitants (UN, 2018). Air quality in the SA cities is a major problem. Measurements of particulate matter of 2.5 microns or less ($PM_{2.5}$) are available for 37 cities in these countries, 35 of them (representing approximately 15% of SA population) experience annual averaged concentrations exceeding the level recommended by the World Health Organization (WHO, 2017).

This paper seeks to evaluate, give guidance and provide insight into the differences and similarities among emissions inventories for five selected SA countries, and possible reasons for such discrepancies. This analysis focuses foremost on air pollutants. The global emission inventories and local emission inventories for selected cities in the five SA countries together with the sources considered are presented in section 2. Sections 3 and 4 present a comparative analysis of the estimated emissions between the five countries investigating the reasons for discrepancies. A discussion of the main results is presented in section 5 followed by conclusions in section 6.

2 Methods and data

2.1 General country information and data

South America has an area of 17,840,000 km^2 and its population in 2010 is estimated at 395 Million (UN PD, 2017) (**Table 1**). The five countries selected in this study for analysis of the available emissions data are Argentina, Brazil, Chile, Colombia and Peru. They cover 80% of the land area, and 84% of the population of South America (**Table 1**).

Country	ISO3 code	Area (km^2)[a)]	Total Population (10^6)[b)]	Urban Population [%][c)]
Argentina	ARG	2736690	41.2	92
Brazil	BRA	8459420	196.8	87
Chile	CHL	743800	17	88
Colombia	COL	1109500	45.9	81
Peru	PER	1280000	29.4	78
Sum of 5 countries		14329410	330.3	86
Total South America		17840000	395.3	84

Table 1: Selected generic data of the selected five countries in South America
[a)] Source FAOstat accessed through https://unstats.un.org/
[b)] UN PD (2017)
[c)] UN PD (2018)

The population of the selected countries is estimated at 330 million in 2010. The demographic development of the countries is similar, in all five countries the population doubled since 1970. When we focus on the last 20 years (1995-2015), the population has grown by about 30% and again the pattern for the 5 countries is similar. The development in gross domestic product (GDP), however, has been less similar and less gradual (Figure 1). Since Brazil is by far the largest country, its GDP follows the average rather closely but Colombia and Peru are clearly below the average GDP, while Chile and Argentina are above the average. Moreover, while the GDP for Chile has mostly increased since beginning of the 1980's, Argentina experienced a decrease in GDP from 1998 until 2003 and Peru mostly shows an increase in the last decade. A common feature for all SA countries is the pervasive inequity (e.g., Solimano and Schaper, 2015; PNUD, 2017), which is also relevant when considering consumption, emission and exposure patterns (e.g., Gallardo et al, 2012b; Carpenter and Quispe-Agnoli, 2015).

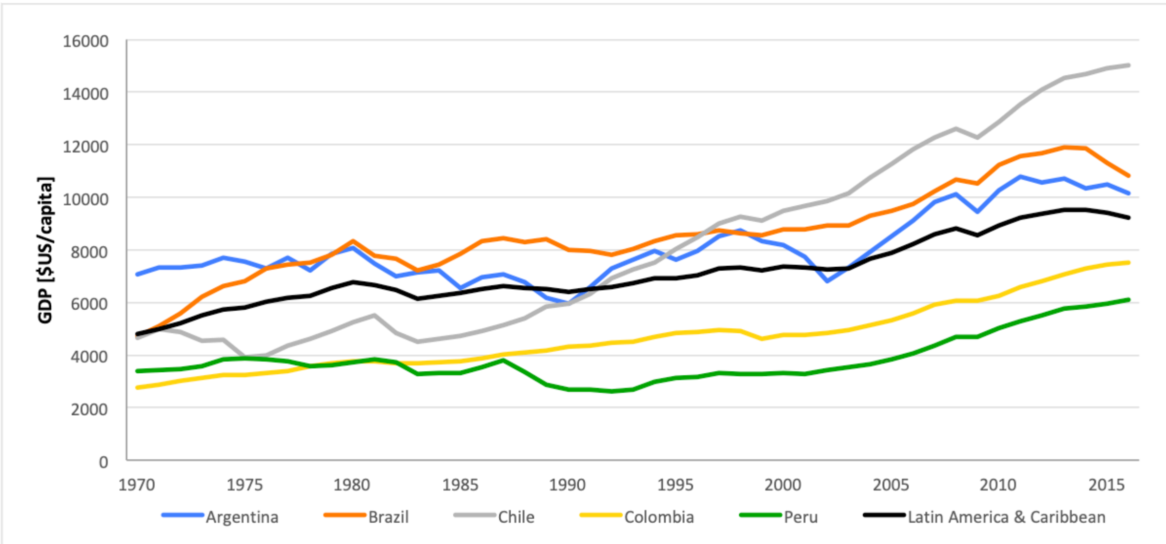

Figure 1: Gross Domestic Product (GDP) per capita ($US/capita in constant 2010 prices) for the same five selected countries in SA. In addition, the GDP corresponding to Latin America and the Carribbean is also included.

2.2 Global emission data

Emission inventories providing data for SA countries from 1970 to present are collected and compared for the pollutants of primary concern: nitrogen oxides (NOx), sulfur dioxide (SO_2), Black Carbon (BC), particulate Organic Carbon (OC), particulate matter of 10 microns or less (PM_{10}), particulate matter of 2.5 microns or less ($PM_{2.5}$), carbon monoxide (CO), and ammonia (NH_3). Methane (CH_4) is important in atmospheric chemistry (e.g., ozone formation) but this pollutant is explicitly included in the GHG emission inventories and is therefore not discussed in this study. A comparison of the data from the various inventories by pollutant and by country for the time period of 1970 to 2010 is given in section 3.1.

Although the total number of (global) emission inventories that provide information on SA is substantial (**Table 2**), a more in-depth analysis is made for a selection of few recent inventories. The selection of inventories is based on;

1. Inventories need to include the recent period 2000-2010 for comparison with other local data sources such as SA city inventories
2. From every “family” of EIs we take the latest version at the time of the start of our investigation; e.g. we look in detail to EDGAR 4.3 and not version 4.2.
3. The inventory needs to have sectorial emissions information.

Based on these criteria our further analysis focuses on EDGAR 4.3.1, ECLIPSE v5, CEDSv3 and MACCity. The MACCity inventory for years after 2000 is a projection based on the RCP8.5 projection by Riahi et al. (2007). As it was used in several studies in support of IPCC AR5 analysis, we include it for reference but will not analyze the implied emission factors or discrepancies in trends compared to the other bottom-up inventories. A brief introduction to the selected inventories is provided below; we refer to the provided references for more details. The emission inventories are accessible through the ECCAD server (https://eccad.aeris-data.fr/)

Acronym	Period	Reference and/or website
MACCity	1980-2010	Granier et al., 2011 http://eccad.aeris-data.fr/
ACCMIP	1980-2010	Lamarque et al., 2010 http://eccad.aeris-data.fr/
RCPs	2000-2010	Van Vuuren et al., 2011 http://www.iiasa.ac.at/web-apps/tnt/RcpDb
EDGAR v4.2	1970-2008	Janssens-Maenhout et al., 2013 http://edgar.jrc.europa.eu/
EDGAR v4.3.1	1970 and 2010	Crippa et al., 2016 http://edgar.jrc.europa.eu/_os
HTAPv2	2008 and 2010	Janssens-Maenhout et al., 2015 http://edgar.jrc.europa.eu/htap_v2
ECLIPSE v4a	2005-2010	Stohl et al., 2015 http://eclipse.nilu.no http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv4a.html

ECLIPSE v5	1990-2010	Klimont et al., 2017 http://eclipse.nilu.no http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5a.html
Junker&Liousse	1860-1997	Junker and Liousse, 2008
PKU	2002-2013	Huang et al., 2014 http://inventory.pku.edu.cn
CEDS	1950-2014	Hoesly et al., 2018 http://www.globalchange.umd.edu/ceds/

Table 2: Overview of selected global emission data sets containing data for anthropogenic emissions for South America.

2.2.1 EDGAR v4.3.1 (January 2016)

The Emissions Database for Global Atmospheric Research (EDGAR) provides global historic anthropogenic emissions of GHG and air pollutants by country, and sector. EDGAR uses a bottom-up methodology with international activity data and emission factors (Janssens-Maenhout et al., 2017; Crippa et al., 2018). The estimated national emissions by sector are distributed on a 0.1ºx0.1º grid using geospatial proxy data.. In EDGARv4.3.1 emissions are calculated for gaseous and particulate air pollutants per sector and country for the time series 1970-2010. This version v4.3.1 is the official release of the EDGAR database used for PEGASOS scenarios (Crippa et al., 2016). Source sector specification follows the IPCC 1996 code, as was also done for EDGAR v4.2).

2.2.2 Eclipsev5

The ECLIPSE emission data set was created with the GAINS (Greenhouse gas – Air pollution Interactions and Synergies; http://gains.iiasa.ac.at) model (Amann et al., 2011), which calculates emissions of air pollutants and Kyoto greenhouse gases in a consistent framework. The GAINS model holds essential information about key sources of emissions, environmental policies, and further mitigation opportunities for 170 country-regions. The model relies on international and national statistics of activity data for energy use, industrial production, and agricultural activities for which it distinguishes key emission sources and related control measures. Several hundred technologies to control air pollutant and greenhouse gases emissions are represented allowing simulation of implemented air quality legislation. Recently the regional resolution of the global GAINS model has been improved by distinguishing more countries in Latin America where five regions were replaced with

13 regions in version V5a, including most countries of South America (UNEP/CCAC. 2018). For more details we refer to Klimont et al. (2017).

2.2.3 CEDSv3 (as of March 2017)

The Community Emissions Database System (CEDS) is an open source data system that produces global, historical (1750 - present) estimates of anthropogenic acidifying gases (Hoesly et al., 2018). Emissions are estimated annually and resolved by country, sector, and fuel, and then gridded by year and sector with monthly seasonality. CEDS estimates rely on existing energy consumption data sets and regional and country-specific inventories to produce emission trends over recent decades. A historical emissions dataset was released in 2016 for use in research, including CMIP6. For more detailed information see www.globalchange.umd.edu/CEDS

2.2.4 MACCity

The MACCity emissions dataset is based on the ACCMIP (Atmospheric Chemistry and Climate - Model Intercomparison Project) historical emissions dataset developed by Lamarque et al. (2010) and combined with the IPCC AR5 future emissions scenarios called RCPs (Representation Concentration Pathways, Van Vuuren et al., 2011). The ACCMIP and the RCPs emissions dataset have been adapted and extended on a yearly basis for the period 1990-2010. Anthropogenic emissions were interpolated on a yearly basis between the base years 1990, 2000, 2005 and 2010. For the years 2005 and 2010, the RCP 8.5 emissions scenario was chosen. For biomass burning emissions, the ACCMIP dataset was extended as well on a monthly basis.. Best reference to the data set is Lamarque et al (2010).

2.3 City emission data in South America

Local emission inventories are compiled for megacities in South America to address deteriorated air quality and implement measures to reduce concentration of air pollutants in these cities (D’Angiola et al., 2010; Gallardo et al., 2012a).

For each of the five countries, one city was selected to evaluate the local inventory against emissions from down-scaled global inventories. A city inventory contains the emissions occurring within the territory of the city (Table 3) and therefore the corresponding emissions from global inventories are extracted based on the squared area surrounding each city. To assess the impact of the choice of the area surrounding the city, two domains are considered when extracting the city emissions from global inventories; a small relatively tight domain following the city limits and a somewhat larger one to test if potential sources that in fact belong to the city are not included in the smaller domain. This method is rather crude and may introduce errors in the estimate, which are discussed in section 4.

City	Country	Year (s)	Sectors	Pollutants	Reference
Buenos Aires	Argentina	1970-2012	Transport	PM_{10}, NO_x, SO_2, CO, VOC	D'Angiola, et al., 2010
Bogota	Colombia	2012	Transport & Industry	PM_{10}, NO_x, SO_x, CO, CO_2, VOC	Rojas & Peñalosa, 2012; Pachón et al., 2018
Lima	Peru	2014	Transport, Residential, Industry	PM2.5, PM_{10}, NO_x, SO_x, CO, CO_2, VOC	Reátegui et al., 2018
Santiago	Chile	2012	Transport, Industry, Residential, Agriculture, Construction, Total	PM2.5, PM_{10}, NO_x, SO_x, CO, CO_2, VOC, CH_4, NH_3	USACH, 2014
Rio de Janeiro	Brazil	2013	Transport	TSP, NO_x, SO_x, CO	GQA, 2016

Table 3: Overview of cities data (city, year(s), pollutants, reference) – see section 3.1 (Please add reference to each inventory)

2.4 Source sector definitions

The various global inventories used in this paper do not all share the same source sector definitions. Therefore, we have aggregated several sectors to come to a common sector breakdown (**Table 4**). This implies that individual inventories may have a more detailed sector breakdown than we use here. For example, EDGAR provides emission data for 12 IPCC 1996 main source categories whereas we aggregate the data into 8 different sectors.

Source sector	Description
Agriculture	Agriculture
Power	Power generation, transformation industry and refineries
Industry	Combustion and process emissions from industry
FF_prod	Fossil Fuel production, exploration and transmission
Residential Combustion	Residential and small commercial stationary combustion
Waste	Solid waste disposal and waste incineration (without energy/heat production)
Transportation	Transport excluding international shipping and excluding international non-LTO[a)] aviation
Solvents	Use of solvents

Table 4: The aggregated source sectors used for intercomparison of selected inventories and city inventories. Note that in this work, Agriculture does not include savanna burning, but does include agricultural waste burning. Furthermore, CEDS includes agriculture waste burning under Waste – not under agriculture; ECLIPSE includes flaring emissions.

[a)] Landing and take off

3 Results

3.1 Emission inventories by pollutant and by country

The SO_2 and BC emissions from the different global emission inventories (**Table 2**) for the five SA countries are plotted as a function of time (Fig 2, Fig 3, respectively). Similar figures for other pollutants are presented in Supplement material section 1 (Figures S01 to S06). The main observation is that emission estimates from different inventories widely vary and do not show a trend to converge. For some countries the early years vary widely (e.g. Argentina SO_2), while for others the discrepancy increases with time (e.g. Peru SO_2). Overall, discrepancies of a factor 2-3 are present for all countries and pollutants. Some of the inventories shown in **Table 2** and Fig 2 and Fig. 3 are no longer considered up to date but the overall overview is useful because these inventories were used in important global assessments and it is not unlikely that those results may be sensitive to such differences in emission data. We will not discuss all these discrepancies in detail. In general, it can be stated that discrepancies between the emission inventories have multiple reasons including:

1. Emission factors may have been updated and improved over time.
2. Activity data may have been updated and improved over time.
3. New sources may have been identified over time.
4. The definition of which sources to be included in the inventory may differ between inventories.

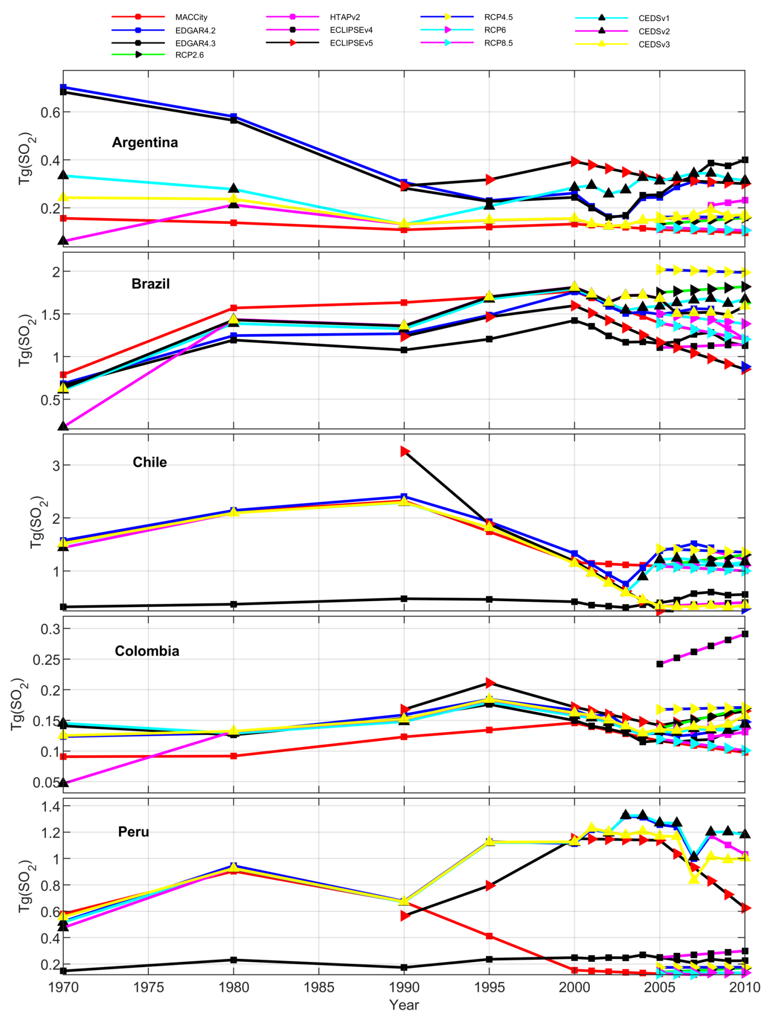

Figure 2: Total annual sulphur dioxide (SO_2) emissions [Tg/yr] for the period 1970 to 2010 for global emission inventories: MACCity, EDGARv2, EDGARv3, HTAPv2, ECLIPSEv4, ECLIPSEv5, RCP2.6, RCP4.5, RCP6, RCP8.5, CEDSv1, CEDSv2 and CEDSv3. See figure legend for colours and symbols associated to each inventory.

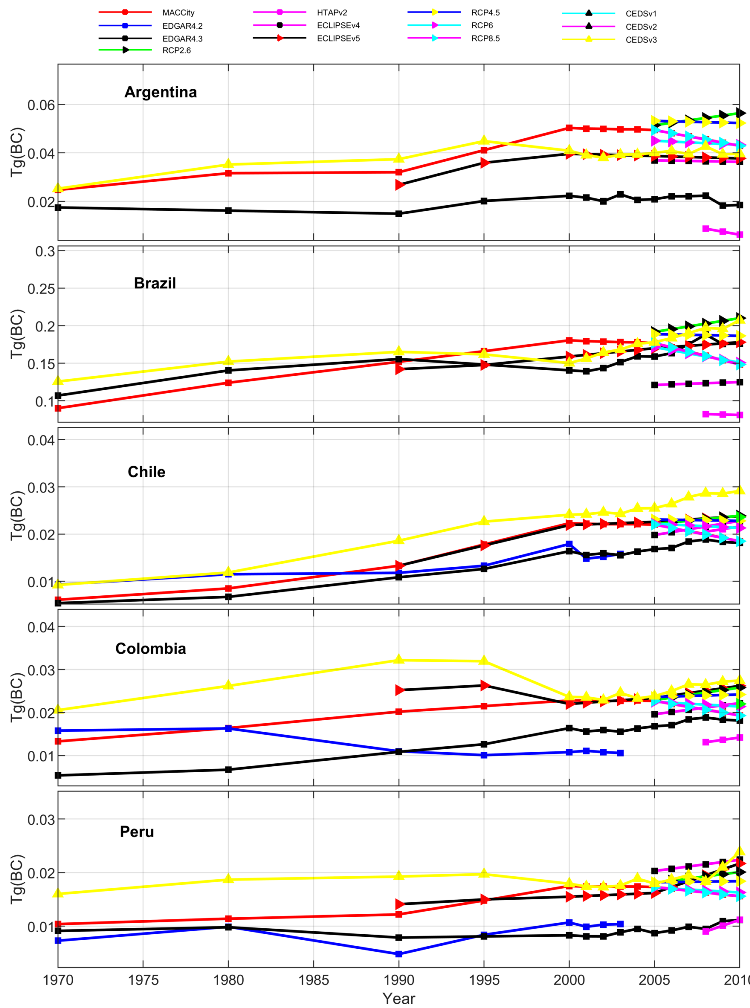

Figure 3: Same as Figure 2 but for black carbon (BC) and global inventories: MACCity, PKU, Junker-Liousse, HTAPv2, ECLIPSEv4, ECLIPSEv5, RCP2.6, RCP4.5, RCP6, RCP8.5, EDGARv3 and CEDSv3.

Based on the criteria outlined in section 2.3 we focus a further analysis on 4 recent emission inventories; EDGAR 4.3.1, ECLIPSEv5, CEDSv3 and MACCity and restrict the analysis only to the years 1995 to 2010. Subsequent versions of emission inventories can differ substantially because of some of the reasons outlined above. Compare for example Eclipse v4 and v5; CEDSv1 and v3 in Figure 2 and Figure 3. Klimont et al. (2017) document that the major change between their ECLIPSE v5 and previous ECLIPSE versions is that IEA and FAO statistical data were reimported for the period 1990-2010, international shipping was included and that their global BC numbers are higher than previously published owing primarily to inclusion of new sources. Another, often overlooked issue is the inclusion or exclusion of semi-anthropogenic sources like forest fires or agricultural waste burning. Compare for example HTAPv2.2 and EDGARv4.3.1 in Figure 3 for Brazil. The emissions for all source sectors are equal but HTAPv2.2 excludes agricultural waste burning, and EDGARv4.3 includes it. In SA this is a relevant source and therefore HTAPv2.2 and EDGARv4.3.1 appear different inventories but the difference is simply a result of inclusion or exclusion of agricultural waste burning. Out of simplicity, the version of each inventory will be omitted henceforth and thus inventories will be referred to as EDGAR, ECLIPSE and CEDS.

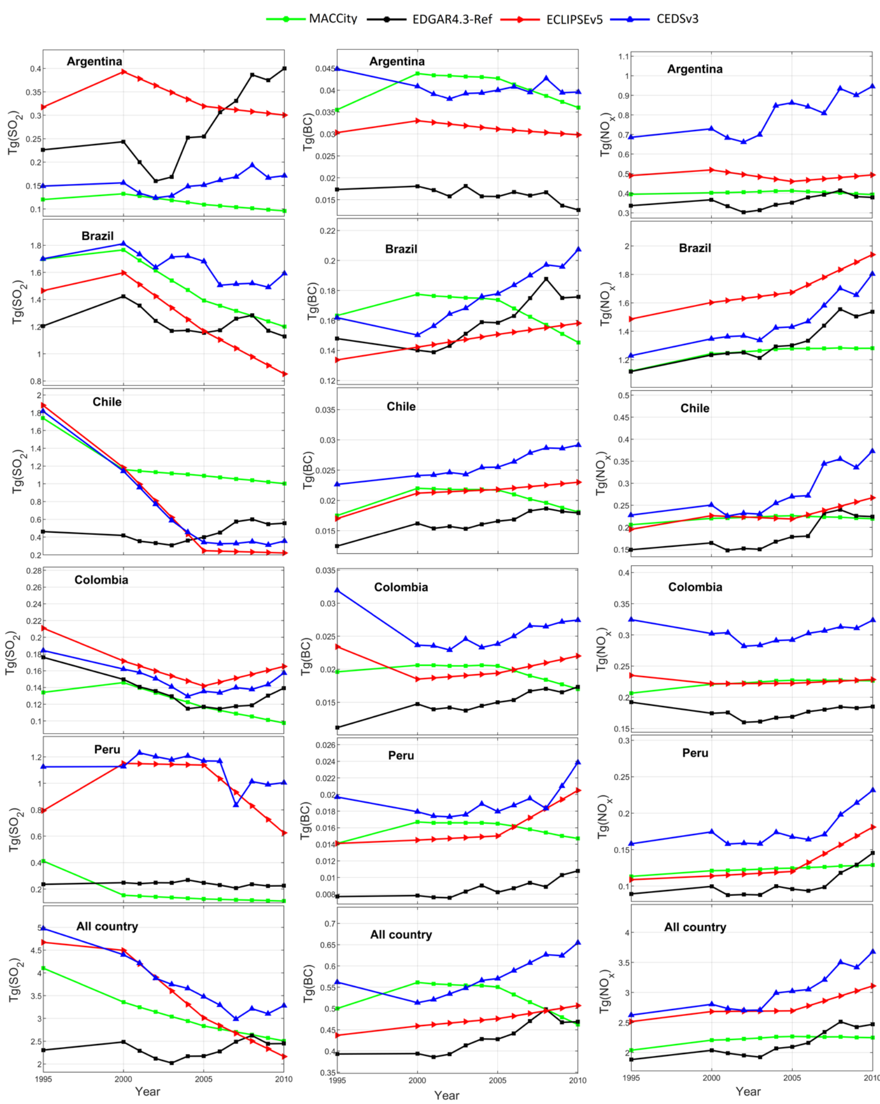

Figure 4: Total annual emissions of SO_2 (left), BC (centre) and NO_x (right) [Tg/yr] for global inventories MACCity (green), EDGAR4.3 (black), ECLIPSEv5 (red) and CEDSv3 (blue) for the period 1995 to 2010.

It is also important to note that emission inventories may show comparable patterns for one country and significantly diverse patterns for another country. Differences are seen in terms of trend and/or magnitude and this will be mostly related to different key source contributions in different countries. For SO_2 these differences are seen both in terms of trends for Argentina and magnitude for Colombia and Peru (Figure 4). Whereas for NOx and BC, differences are mostly in terms of magnitude; see for example Argentina and Colombia for NOx and Brazil and Chile for BC (Figure 4) (See figures S7 to S10 for equivalent figures of the other pollutants considered in this work). We highlight that for most species the SA year-to-year variability (lower panels) is dominated by emissions in Brazil except for SO_2 and NOx. For SO_2 annual variability is also determined by emissions in Chile and Peru resulting from their mining activities. Argentina also contributes to the SA year-to-year variability for NO_x emissions.

In addition to the spread in total emissions presented above, the EIs also differ in the relative contribution of each sector (Table S1 in supplement material). While the diversity between the emission inventories is small for SO_2 and NH_3 it is considerable for the other species. A more detailed description of this diversity in sectorial emissions between inventories is provided in the supplement material.

3.2 Particulate matter emissions

Particulate matter and its components, BC and OC are important species for air quality, health and climate change (Bond et al, 2013; IPCC, 2018). Next to BC and OC, $PM_{2.5}$ and/or PM_{10} can contain other components like sulphates, nitrates and mineral particles but in combustion emissions the bulk is BC + OC. Therefore, looking at BC+OC in relation to PM provides information on the type of emission factors used. Moreover, theoretically BC+OC cannot exceed $PM_{2.5}$ as BC and OC emissions are smaller than 2.5 μm. The contribution of BC and BC+OC to $PM_{2.5}$ as well as the contribution of $PM_{2.5}$ to PM_{10} is analysed in more detail in Table 5. In addition, we pay attention to the ratio of CO to NOx (Table 5), which may provide insights into the role of traffic emissions (Monks et al, 2015; Gallardo et al, 2012a; Vivanco and Andrade, 2006). Note that the present analysis and Table 5 are only for Transport emissions.

EDGAR has in general larger BC content in $PM_{2.5}$ than ECLIPSE except for Chile. In both inventories this contribution increases from 1995 to 2010 except for Argentina, and this increase is larger in ECLIPSE than EDGAR. Although BC is the product of incomplete combustion, such a trend can be explained by better combustion at higher temperatures, which first causes less particulate OC formation, and in a later, further advanced stage may also reduce the formation of BC. For the fraction of BC+OC in $PM_{2.5}$, EDGAR presents the same features as for BC; decreasing trend in Argentina and increasing one in the other countries and the largest increase in Chile. This similarity between both ratios is likely due to the use of the same drivers to estimate the emissions. However, ECLIPSE does not seem to use the same drivers to estimate BC and OC emissions; while for the ratio $BC/PM_{2.5}$ the country with the largest growth was Colombia, for the ratio $(BC + OC)/PM_{2.5}$ Chile and Peru presented the largest increase. We note that both, Argentina and Peru, have unrealistic fractions larger than 1 in EDGAR (Table 5)[1] The use of independent OC emission factors in EDGAR (that is, independent from $PM_{2.5}$) could explain these unrealistic values and allow BC+OC to be larger than $PM_{2.5}$. In addition, EDGAR considers that all emitted PM_{10} for road transport is also $PM_{2.5}$ while ECLIPSE estimates the coarse PM fraction (PM_{10}-$PM_{2.5}$) to be approximately 10%, and gradually increasing over time from approximately 8% to 11%. Finally, the CO/NOx ratio decreases over time with a comparable decrease in both inventories. When comparing the ratios CO/NOx by country and across the years we see a consistent lower CO/NOx ratio for ECLIPSE compared to EDGAR. Yet, while in EDGAR, both Argentina and Chile show an increasing trend since 2005; in ECLIPSE only Chile presents this trend. The CO/NOx emission ratios are important for atmospheric chemistry and composition. If better national data would be available, this discrepancy should be analysed and reconciled or at least better understood as it highlights a fundamental difference.

[1] This is now corrected in version EDGARv4.3.2 where the PM2.5biofuel, PM2.5fossil and BC and OC have been revised and a ratio smaller than 1 is guaranteed for the sum of BC and OC over the sum of PM2.5fossil and PM2.5biofuel.

Transp.	EDGAR4.3.1-Ref			ECLIPSEv5			EDGAR4.3.1-Ref	ECLIPSEv5
1995	**BC/PM2.5**	**(BC+OC)/ PM2.5**	**PM2.5/PM10**	**BC/PM2.5**	**(BC+OC)/ PM2.5**	**PM2.5/PM10**	**CO/NOx**	**CO/NOx**
Argentina	0.71	1.18	1.00	0.45	0.82	0.94	14.28	5.51
Brazil	0.52	0.86	1.00	0.45	0.82	0.93	9.57	6.31
Chile	0.38	0.63	1.00	0.44	0.84	0.91	16.77	7.76
Colombia	0.48	0.80	1.00	0.20	0.78	0.91	28.71	12.26
Peru	0.65	1.08	1.00	0.36	0.81	0.92	12.57	5.88
2000								
Argentina	0.69	1.14	1.00	0.48	0.84	0.94	9.70	4.01
Brazil	0.53	0.89	1.00	0.48	0.84	0.92	8.86	5.51
Chile	0.51	0.81	1.00	0.48	0.87	0.90	17.87	6.76
Colombia	0.48	0.79	1.00	0.33	0.80	0.89	23.14	10.17
Peru	0.63	1.04	1.00	0.44	0.85	0.92	10.43	5.46
2005								
Argentina	0.67	1.09	1.00	0.50	0.83	0.93	8.16	3.28
Brazil	0.50	0.83	1.00	0.50	0.84	0.91	7.22	4.81
Chile	0.46	0.73	1.00	0.54	0.90	0.89	11.36	5.08
Colombia	0.52	0.84	1.00	0.39	0.81	0.89	17.66	8.13
Peru	0.71	1.15	1.00	0.48	0.86	0.91	9.27	4.55
2010								
Argentina	0.69	1.13	1.00	0.53	0.85	0.92	13.02	3.13
Brazil	0.54	0.88	1.00	0.53	0.87	0.90	6.11	3.56
Chile	0.50	0.79	1.00	0.57	0.93	0.86	12.01	5.42
Colombia	0.52	0.84	1.00	0.46	0.82	0.89	11.51	5.88
Peru	0.84	1.32	1.00	0.51	0.88	0.89	7.52	3.74

Table 5: Road transport emission fraction of BC in PM2.5, BC+OC in PM2.5, fraction of PM2.5 in PM10 and CO/NOx ratio for EDGAR and ECLIPSE.
Note: the color scale only applies to CO/NOx one can see that it becomes lighter from 1995 to 2010 and that EDGAR CO/NOx ratio tends to be higher than ECLIPSE

3.3 Comparison of city emission data with global inventories

Emission estimates from local and global inventories are intercompared for total emission as well as individual source sectors (Transport, Industry and Residential) whenever available. To keep the comparison transparent only EDGAR and ECLIPSE are considered in this analysis because CEDS partly relies on EDGAR and other inventories. One city per country was selected; namely Bogota, Buenos Aires, Lima, Rio de Janeiro and Santiago (Table 3). Unfortunately, the local city inventories differ substantially in terms of sectors included, period and pollutants considered (Table 3). Furthermore, while both global EIs are available for the year 2010, the local city EIs are only available for specific years and the year nearest to 2010 was selected (Table 3). The corresponding emission from the global EIs for a given city was derived by considering emissions within a squared domain surrounding the city. To assess the sensitivity of the method, two domains are considered; a small one with its limits in the vicinity of the city and a larger one considering potential sources excluded in the smaller domain. Maps illustrating each domain as well as the emissions included in each one are provided in the supplement material (Figure S11 to S13).

The total emissions from Santiago and Buenos Aires (the only two cities with emission estimates for all sectors) will be analyzed first and then the analysis will be extended to sectorial emissions for the remaining cities. We focus on two pollutants; NO_x and PM_{10}, since the goal of this analysis is not to validate either inventory but to highlight differences between them and identify the impact of local information in the estimate. In **Table 6** an overview is given for the cut-out from EDGAR and ECLIPSE that represents the Santiago and Buenos Aires metropolitan areas by sector and how it relates to national total emissions. Knowing that over 1/3 of Argentina's and Chile's population live in their corresponding capitals (Buenos Aires and Santiago, respectively) their share of Transport emissions appears small but an in-depth analysis by inventory would be needed to fully understand this. Furthermore, regardless of the domain considered, ECLIPSE attributes a larger fraction of the total national transport emissions to both cities than EDGAR while the opposite occurs for industrial emissions. For residential and total emissions, percentages between both inventories and for both domains are comparable. In terms of NOx emissions, for Buenos Aires both inventories assign comparable percentages of the total emissions to transport but differ for residential and industrial emissions. In spite of these differences, both inventories allocate comparable fractions of the national total emissions to Buenos Aires. On the contrary, for Santiago both inventories present comparable percentages for transport and industrial emissions but differ in residential and total emissions. Detailed, spatial explicit national inventories for Argentina and Chile would be extremely useful to elucidate the reasons for the discrepancies highlighted above. For EDGAR the geospatial proxy data have been disclosed in Janssens-Maenhout et al. (2017) for the sake of transparency but exactly why higher or lower shares of emissions are attributed to the city remains unclear.

The relative magnitude of total NOx emissions for both cities depend on the domain or regions considered to estimate the city emission from global inventories. EDGAR presents for Buenos Aires comparable emissions to the local inventory when the smaller domain is considered and larger emissions when using the larger domain, whereas the contrary is observed for Santiago where the estimate based on the larger domain is comparable to the local inventory and the one based on the smaller domain presents smaller emissions than the local one (Figure 5). Spatial distribution of the emission in each city explains this opposite behavior; while for Buenos Aires most of the city NOx emissions are distributed within the city domain enclosed by the smaller domain, for Santiago the emissions are distributed beyond the city limits and fall thus outside of the smaller domain but are captured by the larger one (Figure S12e and S12n in the supplement material). Contrary to EDGAR for Santiago, ECLIPSE presents larger emissions than the local inventory regardless of the domain used to estimate the city emissions. For Buenos Aires however, the estimate based on the larger domain is comparable to the local inventory while the estimate based on the smaller domain is smaller than the local inventory. The local inventories in both cities estimate smaller emissions for the Transport sector (Figure 5). The contrasting estimates between Transport sector and the Residential and Industrial sectors between the Global EIs and the local city EIs may be partly related to the distribution proxies used in the global inventories but the impact on total emissions attributed to the cities is large (Figure 5).

Except for Transport, both EIs (and regardless of the domain used to estimate the city emissions) present larger PM10 emissions than local inventories for both cities. Finally, for total emissions both inventories present comparable emissions for Buenos Aires whereas for Santiago EDGAR presents larger emissions than ECLIPSE per domain.

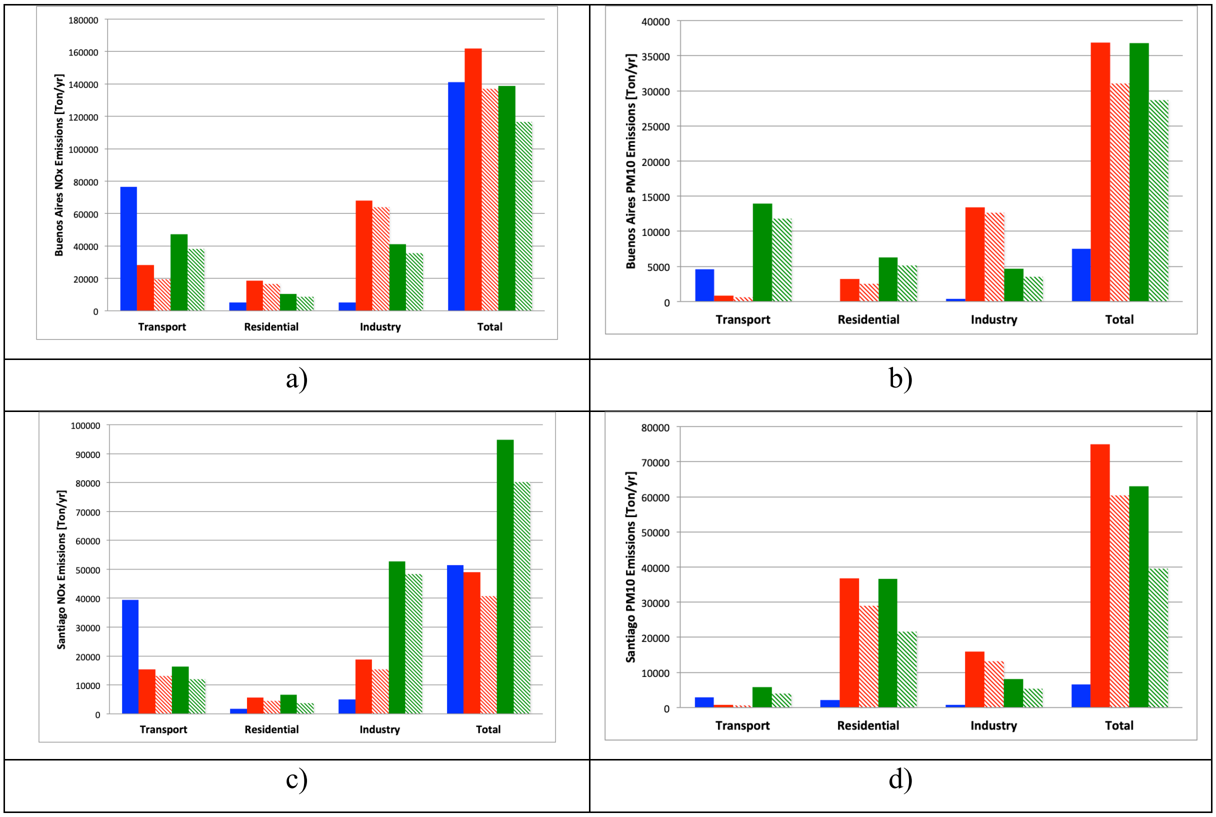

Figure 5: Total and sector emissions (Transport, Residential and Industry) of NOx and PM10 for Buenos Aires (a and b, respectively) and Santiago (c and d, respectively) from local EIs (blue) as well as global EIs EDGAR (red) and ECLIPSE (green). Estimates for a large (filled) and small (shaded) domain are included for each global EI. See figure S01 for limits of each domain and city boundaries.

The analysis is extended to all selected cities for the Industrial, Residential and Transport sector. Emissions from both global inventories are presented only when a local inventory is available for comparison (Figure S14 in supplement material). The analysis focuses on cities not analyzed before, namely Bogota and Lima. Residential and Industrial emissions present the same features for both pollutants; global inventories are mostly larger than the local ones. While in general EDGAR emissions are larger than ECLIPSE ones for both sectors in Bogota the opposite is seen for Lima where ECLIPSE is larger than EDGAR. We note that for Lima, estimates for each inventory between both approaches are mostly the same indicating that allocation of the sources are confined within the city included in the smaller domain. The opposite of Residential and Industrial emissions is seen in the Transport sector for NOx where the local inventories are mostly larger than the global ones (probably due to the geospatial allocation of roads x population into the city).

Exception to this is Rio de Janeiro where based on the domain considered for the estimate the estimates are either larger (EDGAR based no large domain), smaller (ECLIPSE based on small domain) or comparable to the local inventory. For PM10 emissions from the Transport sector, EDGAR mostly estimates smaller emissions than the local inventory whereas ECLIPSE in general estimates larger emissions with the exception of Lima where they are comparable with the local inventory.

Although the magnitude of the estimated emissions from the global inventories for each city depends on the region considered, the results with respect to the local inventories are in general independent of the selected domain.

	EDGAR				**ECLIPSE**			
	Transport	RCO	Industry	**All sectors**	Transport	RCO	Industry	**All sectors**
PM10 (tons/yr)								
Buenos Aires	580 (868)	2491 (3192)	12643 (13424)	**31020 (36836)**	11805 (13943)	5146 (6295)	3553 (4647)	**28655 (36814)**
Percentage of national total	6% (8%)	26% (33%)	28% (30%)	**8% (9%)**	24% (29%)	31% (38%)	8% (11%)	**11% (15%)**
Santiago	618 (751)	28976 (36834)	13170 (15989)	**60396 (74994)**	3994 (5842)	21635 (36640)	5398 (8182)	**39611 (62920)**
Percentage of national total	4% (5%)	31% (39%)	38% (47%)	**25% (31%)**	27% (39%)	25% (43%)	10% (15%)	**21% (34%)**
NOx (tons/yr)								
Buenos Aires	19386 (28285)	16664 (18545)	64053 (68106)	137160 (161879)	38348 (47182)	8760 (10442)	35508 (41205)	116671 (138942)
Percentage of national total	10% (15%)	68% (75%)	75% (80%)	29% (34%)	12% (14%)	45% (54%)	51% (59%)	23% (28%)
Santiago	13125 (15299)	4420 (5564)	15398 (18769)	40774 (49008)	12005 (16360)	3644 (6546)	48420 (52740)	80195 (94801)
Percentage of national total	13% (15%)	51% (64)%	47% (57%)	17% (21)%	9% (13%)	39% (70%)	53% (58%)	30% (35%)

Table 6: Extracted city domain emissions of small (large) domain for Buenos Aires and Santiago for PM10 and NOx in 2010 and the fraction of national total emissions located in the city domain for 2010

4 Discussion.

Brazil is the largest emitter of all analyzed species, except for SO_2 where its emissions are comparable with those from Chile and Peru. This is not surprising as it is the largest and most populated country in SA. However, when emissions are considered per capita, i.e. they are normalized by population, this domination of Brazil disappears (Figure

6 and 7). To limit the amount of data, we only present normalized data for the inventories ECLIPSE and EDGAR, including more inventories would not give significantly different insights. We only normalize sectors by population if population has a direct link with activities causing emissions, such as heating of houses (residential), commuting (transport) and industrial heat/power use. For sectors like agriculture and industry, population is not a good proxy to normalize emissions. Normalization by population of EDGAR emissions reveals that Chile is, in general, the country with the largest emissions per capita among these five countries. Chile stands out as the largest emitter per capita for SO_2, PM_{10} and OC mostly related to the residential sector and power generation (Figure 6). For BC, Chile is also dominating due to transport and residential emissions. Finally, emissions of CO and NO_x are dominated by the transport sector where again Chile has the largest emissions per capita, but those from Argentina and Brazil follow with smaller differences than for the other species. Normalization of ECLIPSE emissions by population presents similar figures as EDGAR for the power and residential sector with larger implied emission factors (EF) for EDGAR in the power sector but in general, smaller ones in the residential sector (Figure 7). Largest differences between both inventories are seen in the transport sector. While Chile is the largest emitter per capita for PM_{10}, BC, OC and NO_x according to EDGAR, the largest emitter according to ECLIPSE is Argentina. Part of the differences can be understood from geographical data. Chile is the country with a substantial amount of the population living in the South with cold winters and residential emission will be much higher in a cold climate, hence higher per capita emissions for residential sector. Another explaining factor is the economic development, in a relatively affluent country like Chile (see Fig 1) more people will own a car or have access to transportation and as a result more vehicle km (vkm) and transport emissions per person can be expected. This, however, already becomes more difficult to generalize because a more affluent country will have a more modern car fleet with less emission per vkm, but more vkm still create more emissions. So, while per capita transport emissions in Chile rank high (Figure 6 and 7), per vkm they would probably be lower than some of the other countries. The data for power are most difficult to interpret. For SO_2 it is related to the S content in the fuel used but more important we may have to correct for (small) industrial power use and we lack the data to properly do this. Also climatic differences may play a role. In general the normalization in

Figure 6 and 7 suggests we may expect that per capita emissions from Colombia and Peru will still grow.

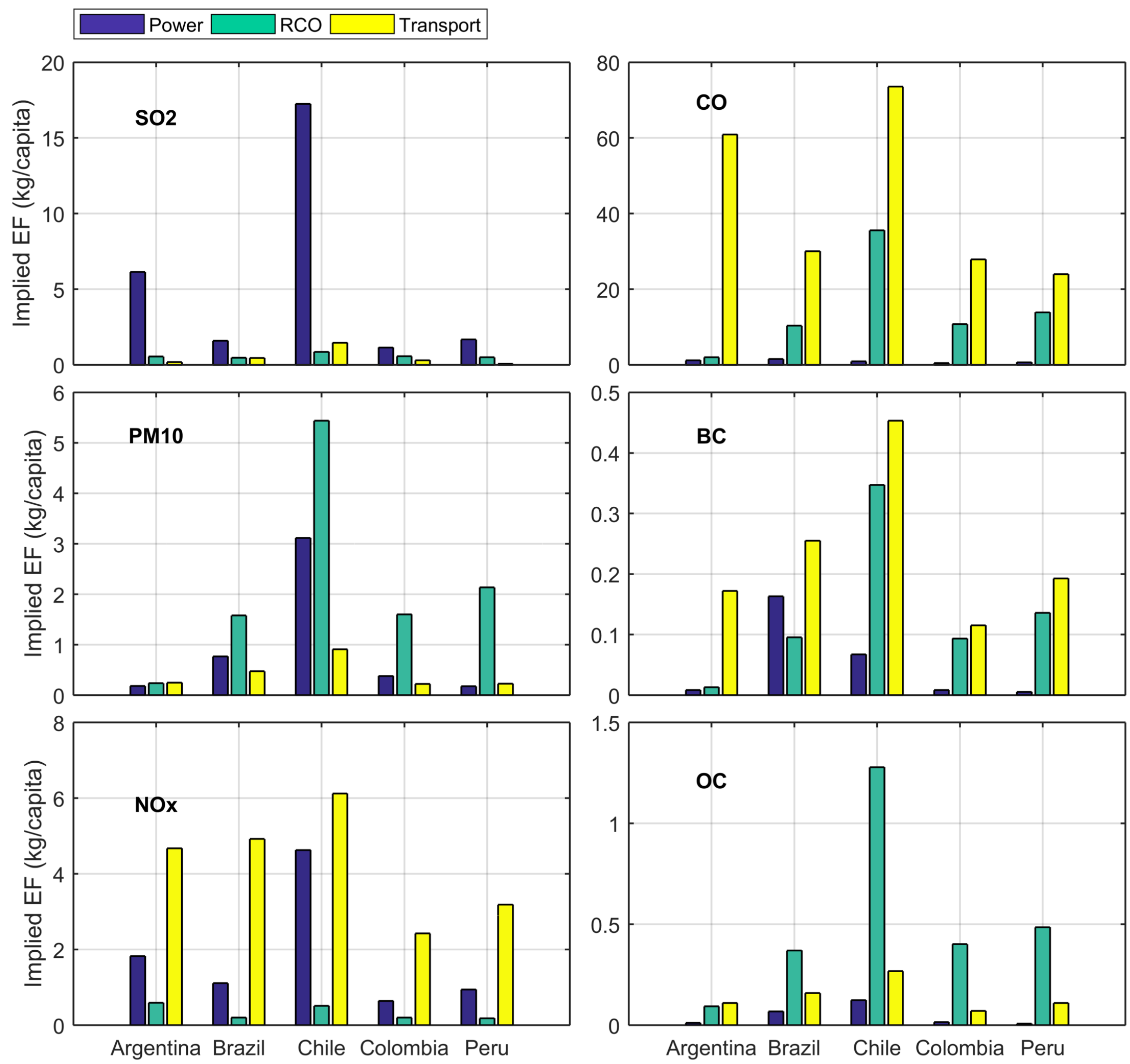

Figure 6: Normalized (by population) national emissions of SO_2, CO, PM10, BC, NOx and OC from the global inventory EDGAR for Transport (yellow), RCO (green) and Power (blue) sector for the five selected countries (Argentina, Brazil, Chile, Colombia and Peru).

Global Inventories use generic statistical country data to estimate emissions by combining for example transportation fuel sales data with emissions factors (e.g. Klimont et al. 2017, Crippa et al., 2018). Using road transport as an example, it is known that he emission factors are strongly dependent on the engine technology, type of fuel and fuel quality. The latter is highly variable and rapidly changing in SA. Global scale inventories try to capture this change over time but can only do so in a rather generic way, at best for individual countries but often by grouping multiple countries in similar “stage of technology development” classes. However, for air quality modelling and exposure in major cities the patterns in, and within, individual countries are important. This is illustrated with diesel fuel in Argentina as an example (Table 7). Argentina has three grades of diesel; Grade 1 (Agrodiesel or Gasoil Agro), is intended mainly for agricultural equipment. Grade 2 (Gasoil Común; common diesel fuel), is intended for the bulk of diesel fuelled vehicles and the sulphur limit of 500 ppm is not strictly enforced. Grade 3 diesel fuel, also known as Gasoil Ultra, is the highest quality diesel fuel. Another complication is that a policy or resolution may be accepted but the delay in implementation can be substantial; as seen in Table 7; a resolution for low sulphur fuels was accepted in 2016 but availability on the market is not expected until 3 years later. This kind of details are important but often only known by local or national experts. In the supplement material we provide additional information on sulphur contents in fuel and its evolution in SA (see Table S2), illustrating the importance of national and subnational policies. However, from **Table 7** we can already see how challenging proper incorporation and spatial distribution will be for global emission inventories. Various grades may exist next to each other and will be implemented differently across the country, severely affecting the emissions in urban population centers. The example of the complex temporal and spatial implementation of the sulphur fuel standards shows the importance of working with local and national teams, especially when high resolution and spatial distribution is important to predict exposure and define mitigation measures. For global scale studies these details will be of limited importance but when understanding the air pollution in South American cities or regions it may be crucial. Also, when assessing climate impacts and mitigation options this may become increasingly important.

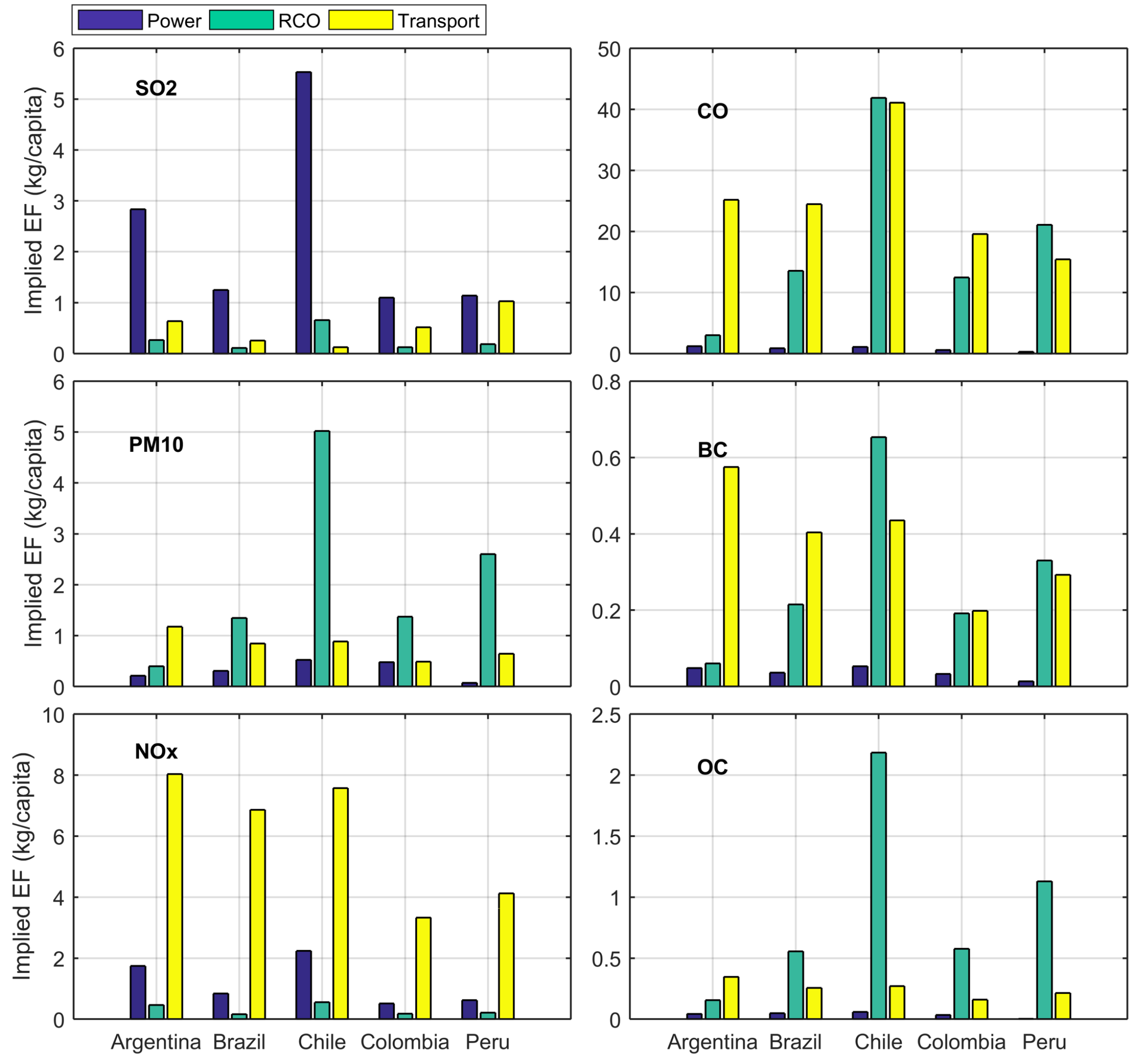

Figure 7: Same as Figure 6 but for global inventory ECLIPSE.

Grade	name	Sulphur limit (ppm)				
		2006	2008	2009	Current	June 2016[a]
1	Agrodiesel or Gasoil Agro	3000	2500	2000	1500	1000
2	Gasoil Comun	1500[b]/2500	1500[b]/2000	500[b]/2000	500	30
3	Gasoil Ultra	500	500	50	10	10

Table 7: Stepwise changes in diesel Sulphur limits in Argentina since 2006.
[a] Date of resolution, market availability is expected in 2019.
[b] Sold in high population zones (more than 90,000 inhabitants, since 2008 50,000 inhabitants)

The analysis of local city emission estimates against results from global inventories for the corresponding domain strongly highlight the importance of including local information. The emission estimates are dramatically different. If emissions are to be applied to forecast air quality and/or to develop mitigation measures the results from down-scaled global inventories as compared to local city data will be entirely different. To come to more general conclusions we have calculated the ratio by source sector for the down-scaled emissions from the global inventories and the local city inventories (Table 8*)*. In this table a value smaller than 1 indicates that the local estimate is higher (green shading), a value higher than 1 indicates the down-scaled estimate is higher (orange shading). The sectors industry and power had to be grouped because in the city inventories this is often one category. A comparison for total emissions could only be made for Santiago and Buenos Aires because other city inventories are not complete. However, for these two cities the sectors transportation, RCO and Industry and power represent 62 ± 10% and 79 ± 9% of the emissions for PM10 and NOx, respectively. Hence, the comparison of the other cities is likely to cover at least the most important sectors and emissions.

Except for Lima, for each global inventory the magnitude of the estimated emission between the two domains (large and small) is mostly different, yet the relative magnitude to the local inventory is in general independent of the selected domain with the exception of total NOx Emissions in Buenos Aires from EDGAR and transport PM10 emissions in Bogota and NOx emissions in Lima from ECLIPSE. Therefore, the following analysis will focus on the dominant features between the local inventory and the equivalent estimate from each global inventory.

The result show that transportation emissions are estimated to be higher in local city inventories in most of the cities, except for Rio de Janeiro where global inventories are higher than the local inventory. Sometimes the result is close to 1 (e.g. PM10 ECLIPSE for Lima) but more often the discrepancies are large up to a factor of 5 (e.g. NOx ECLIPSE for Santiago) or even up to a factor of 10 when the NOx ECLIPSE estimate for Lima based on the small domain is considered. This is remarkable because the selected cities represent a large fraction of the national population and transport emissions are assumed to be in some way proportional to population, and in the case of Santiago 40% of the Chilean population lives in the Santiago Metropolitan region.

City	Inventory	Transport		Residential		Industry		Total	
		PM10	NOx	PM10	NOx	PM10	NOx	PM10	NOx
Bogota	Local	1 [a)]	1	NA[a)]	NA	1	1	NA	NA
	EDGAR-S	0.44	0.19			5.5	3.1		
	EDGAR-L	0.54	0.24			8.1	4.3		
	ECLIPSE-S	0.96	0.07			1.2	3.1		
	ECLIPSE-L	2.25	0.22			3.1	3.8		
Lima	Local	1	1	1	1	1	1	NA	NA
	EDGAR-S	0,30	0.52	49	5.4	8.7	32		
	EDGAR-L	0.35	0.60	50	5.6	8.7	32		
	ECLIPSE-S	1.1	0.17	76	8.2	14.2	39		
	ECLIPSE-L	1.1	0.17	76	8.2	14.2	39		
Rio de Janeiro*	Local	1	1	NA	NA	NA	NA	NA	NA
	EDGAR-S	0.60	1.3						
	EDGAR-L	0.96	1.9						
	ECLIPSE-S	3.0	0.44						
	ECLIPSE-L	5.2	1.1						
Santiago	Local	1	1	1	1	1	1	1	1
	EDGAR-S	0.21	0.33	13	2.5	18	3.1	9.2	0.79
	EDGAR-L	0.26	0.39	17	3.1	22	3.8	11	0.95
	ECLIPSE-S	1.4	0.31	10	2.0	7.3	9.8	6.0	1.6
	ECLIPSE-L	2.0	0.42	17	3.7	11	11	9.6	1.8
Buenos Aires	Local	1	1	1	1	1	1	1	1
	EDGAR-S	0.13	0.25	6.2	3.2	32	12	4.1	0.97
	EDGAR-L	0.19	0.37	8.0	3.6	34	13	4.9	1.2
	ECLIPSE-S	2.6	0.50	13	1.7	8.9	6.7	3.8	0.83
	ECLIPSE-L	3.0	0.62	16	2.0	12	7.7	4.9	0.98

Table 8: Ratio of city domain inventory over local city inventory for PM10 and NOx emissions for transport, RCO and industry & power. A comparison of total emissions could only be done for Santiago and Buenos Aires.

[a)] NA = Not Available; A ratio of 1 for the local inventory indicates the source sector is estimated, if no sector estimate is available in the local inventory, a ratio for EDGAR or ECLIPSE could not be calculated.

*PM as TSP in local

In contrast with the emissions from the transportation sector, the emissions from other sectors (Residential and Industry & Power) are overwhelmingly larger in the downscaled global inventories. The discrepancies can easily amount to a factor 10 or more with an extreme of a factor 70 for ECLIPSE PM10 from the Residential sector in Lima. An overestimation in down-scaled global inventories for RCO emissions is not surprising as they are likely to use population density as a proxy, whereas national or cities information may have better data on which fuels are used where in the country; e.g. more in rural or colder mountainous areas. As a result the ratio for the total emission (Table 8, right column) comes again closer to 1 but the unbalance in individual sectors suggests that this is a case of a better fit for the wrong reasons. Moreover, for air quality modelling emission height and emission timing which vary by source sector are important and for mitigation measures the sector information is crucial. While we highlight that the local information is crucial, it should be acknowledged that this is often lacking (only 2 cities provide a complete inventory) and the use of downscaled global inventories is the only option. A deeper understanding is further hampered by the scarcity of national total emission inventories. For example if the national total emission for transport between EDGAR or ECLIPSE and a national inventory would be similar but the ratio for the major city would be really different, we would know that this is due to spatial distribution and much less likely a fundamental difference in activity and emission factors used. So, national country wide emission inventories are not only needed to model air quality on the national scale but also to place city emissions in perspective. Such an analysis was made by Puliafito et al. (2017) for Argentina by comparing their national inventory (GEAA) with EDGAR. Although the estimated CO_2 emissions were very close (within 1%), large differences were found for the air pollutants with the national inventory being 1.7, 1.1, 1.4 higher for NOx, PM10 and PM2.5 but only 0.4 times the EDGAR estimate for SO_2. In line with our results in Table 8, the attribution to source sectors was often dramatically different partly masking the difference in the totals. Moreover, Puliafito et al. (2017) conclude that the spatial distribution by EDGAR was not adequate for Argentina, especially for transport and residential sectprs, leading to an overestimation in rural areas and an underestimation in urban areas. We refer to Puliafito et al. (2017), for a more detailed analysis.

In addition to the differences of the fraction of national emissions allocated to the different cities in the global inventories, we note that the spatial distribution of these emissions vary largely not only between the inventories but also with respect to the physical location of the actual city (Figure S12 and S13 in the Supplement material). For example, ECLIPSE locates most of the emissions of Santiago outside the city limits extending even over the Andes to the border with Argentina. Similarly, EDGAR although partially distributes the emissions within the city, they are centred on the northern border and an important fraction is outside the city limits.

5 Conclusions

Air quality modeling and exposure assessment needs, next to a validated chemistry transport model, two critical inputs; meteorological data and emissions data. National emission inventories in South America have focused on GHG as part of their obligations to the UNFCC. Only for Argentina a national emission inventory of air pollutants has recently been released (Castesana et al., 2018; Puliafito et al., 2015, 2017). Hence, studies assessing air quality at national, regional or continental scale in SA have been relying on global emission data sets. Several global emission inventories provide estimates for the selected SA countries (Argentina, Brazil, Chile, Colombia and Peru) for the past and present. These have been compiled and compared here with a further analysis focusing on three recent inventories (EDGAR 4.3.1, ECLIPSEv5, and CEDSv3) as they include data at least up to 2010 and can be compared with recent national or city scale emissions estimates. Emissions from these global inventories are compared against available city-scale inventories for a major city in each country by selecting the spatial domain from the gridded data of the global inventory. Large discrepancies are found both between the global datasets as well as when comparing downscaled global emissions data with local/national city emissions data for the same domain. A direct conclusion of these discrepancies is that it is not recommended to use a global emission inventory to derive city emission as input for AQ modelling. The local situation appears to be not properly represented and the results of air quality modelling will significantly depend on the choice of inventory. Moreover a ranking of potential efficient mitigation measures would also depend on the choice of emission inventory. Without more detailed national emission data, a clear conclusion on the origin of discrepancies cannot be made although we have given several suggestions in the discussion. In some cases it is clear that emission factors differ substantially and this may need further attention in the future. At the same time spatial allocation can be another cause for large discrepancies. Simply said, the lack of consistent national inventories prohibits a further analysis. Efforts to create such national gridded inventories in South America are urgently needed.

The emissions analysed in this study, although limited to five countries out of the 12 existing countries in SA, are representative of most of the territory of South America as well as most of its population. This work seeks to increase the understanding of strength and weaknesses of emissions data available for South America by focusing on these five countries. The intention for the future is to include other countries not only from South America but also Central America and the Caribbean.

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