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Air Quality Characterization and Trend Analysis in a Brazilian Industrialized Metropolitan Area in the Period from 1995 to 2022

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Abstract: The Brazilian population grew approximately 9% from 2006 to 2016, and the number of deaths caused by air pollution increased by 14% in Brazil in the same period. Facing the lack of studies on air quality in the Metropolitan Area of Belo Horizonte (MABH)—the third most populous Brazilian metropolitan area—this study aimed to investigate the air quality and the trends of air pollutant concentrations in the MABH between 1995 and 2022, using data from the air quality monitoring network. The methodology consisted of checking MABHs air quality trends following the WHO air quality guidelines. The Mann–Kendall test was used to check statistically for the possibility of tendencies. The results showed a trend of stability in the concentrations of air pollutants in the MABH without any trend of improvement or worsening. However, the time series of the MABH exposed the challenge of ensuring better air quality that protects human health. Furthermore, the results reinforced the importance of focusing on pollutant sources and exposed the need for improvements in air quality management. Thus, it is essential to reverse the current dismantling scenario of the public environmental agencies in Brazil.

Keywords: air quality assessment; air quality monitoring; air quality characterization; air quality trend analysis; air pollution in a Brazilian urban center; Metropolitan Area of Belo Horizonte

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1. Introduction

Human activities have significantly impacted the planet's environment and lifestyle, resulting in air quality issues that pose risks to both human health and the ecosystem [1,2]. A substantial amount of evidence has pointed out the effects of air pollution on human health, particularly in urban centers [2,3]. According to the Global Health Observatory of the World Health Organization, 6.7 million deaths worldwide are attributed to exposure to airborne pollutants [4]. As a result, directing efforts, technological advancements, and financial resources toward air quality research is paramount to comprehending the origin, composition, behavior, interactions, and control mechanisms of air pollution, ultimately safeguarding public health.

Currently, more than 6000 cities across 117 countries employ air quality monitoring programs, showing that approximately 99% of the global population is exposed to dangerously high concentrations of pollutants that exceed the air quality guidelines set forth by the World Health Organization (WHO), thus presenting a grave threat to populations worldwide [5,6]. The worldwide air quality monitoring network is expanding, yielding increasing evidence regarding the consequences of human exposure to air pollutants. According to WHO reports, low- and middle-income countries exhibit higher levels of particulate matter [6]. However, some authors highlighted the challenge of insufficient data for managing air quality and highlighted the importance of identifying the highest local air pollution levels, which is crucial information for assessing global human exposure to air pollution and planning efforts to mitigate its impacts [7].

Implementing efficient instruments and guidelines for controlling and managing local air quality is the initial step to assessing the true state of air quality and identifying external factors influencing it. To establish an effective air quality management program, the following actions are necessary: (i) emission control programs; (ii) installation and operation of air pollution monitoring systems; (iii) assurance of public data accessibility; (iv) dispersion modeling; (v) implementation of more stringent and globally harmonized air quality standards; (vi) local management systems; and (vii) policies encouraging the assessment of air pollution risks to human health [4,7]. Galvão, Feroni, and Orlando [8] clarify how these tools and actions have pros and cons when applied individually, not being able to describe an ideal and complete representation of reality on their own, being such tools mutually complementary.

In Brazil, the population witnessed a growth of approximately 9% from 2006 to 2016, and air pollution-related deaths increased by 14% in the same period [9]. In Brazil, fine particulate concentrations exceeding WHO guidelines [annual concentration of 10 $\mu g/m^3$] were responsible for an increase of 48,700 $\pm$ 7570 deaths due to all causes between 2014 and 2018, highlighting the urgent need to improve air quality policies in Brazil [10].

In 2021, a new edition of the WHO global air quality guidelines was published [11], offering updated recommendations for safeguarding public health from the adverse effects of air pollutants. Additionally, the fact that the final Brazilian standard is outdated, coupled with the country still adhering to the first interim standard with no migration deadline for the subsequent stages, underscores the nation's challenges in effectively monitoring and improving air pollutant concentrations throughout the Brazilian territory.

The air quality monitoring network (AQMN) in Brazil consists of 286 stations, incorporating automatic and manual monitoring, which is insufficient for an appropriate air quality monitoring program. Brazil has approximately 203 million inhabitants and a rate of only 1.4 monitors (one per criteria pollutant) per million inhabitants. As a comparison, for $PM_{2.5}$ monitoring, most European countries and the USA have a rate above 3 monitors per million inhabitants [7]. However, this network is present in only 11 out of the 26 states and the Federal District, with no stations in northern Brazil, despite the region's significant environmental impact due to biomass burning emissions. Furthermore, only 16% of these stations are located outside Brazil's southeastern region, 5.5% in the northeast, 7.5% in the south, and 3% in the center-west region. The southeast of Brazil represents approximately 84% of the national network, with stations in all four states: Rio de Janeiro, São Paulo, Minas Gerais, and Espírito Santo [12]. Consequently, the Brazilian air quality monitoring network lacks representativeness of the national territory, and not all states that monitor air quality can ensure the quantity and quality of the data. According to the Institute of Energy and Environment of São Paulo (IEMA-SP) [12], the main causes are (i) lack of policies and clarity of purpose in the use of air quality data; (ii) scarce human resources; (iii) absence of perennial funding; (iv) lack of strategic planning for the monitoring network; (v) lack of standardization of the data; and (vi) difficulty in publishing data and elaborating reports.

Regarding air quality monitoring in Minas Gerais, the state government adopted the "network and operation by polluting companies" monitoring management model. In this approach, companies that perform potentially polluting activities are responsible for installing and operating monitoring stations based on environmental licensing conditions or terms of conduct adjustments. The environmental agency conducts audits, validates, and publishes the data. The automatic AQMN in the state of Minas Gerais currently comprises 53 automatic stations operating since 1995 [12–14].

The Metropolitan Area of Belo Horizonte (MABH) is the third-most populous Brazilian metropolitan region. Currently, 17 automatic monitoring stations, managed by the State Environmental Foundation of Minas Gerais (FEAM) and funded by polluting industries, continuously operate in the MABH [13]. Some authors have pointed out the existing fragility of the MABHs air quality network, citing limited data availability and a lack of transparency in station data [15–17]. These studies reinforce the need to improve the monitoring network in Minas Gerais and the adoption of more restrictive air quality standards in force in Brazil since current levels do not satisfactorily protect the lives of the population.

Some studies have focused on analyzing air pollution in the MABH, investigating aspects such as (a) evaluation of air quality model results [18,19], (b) evaluation of the influence of meteorology on fine particle concentration [20], (c) identification of some pollutants' behavior and their relation with atmospheric conditions [20], (d) health effects of inhaling fine particulate matter [18,21], and (e) emission inventories and control strategies [19,21–24]. All these studies highlight the need for reliable air quality data for assessment and air quality management. Despite the rapid growth of MABH and the strong influence of industrial and road traffic emissions [18], none of the studies focused on MABH, seeking to know the actual situation regarding air quality and understand the evolution of air pollution in this metropolitan area.

The primary objective of this research is to assess the air quality status in the MABH based on the guidelines set by the WHO. Additionally, this study aims to investigate the temporal trends in atmospheric pollutant concentrations from the inception of automatic air quality monitoring in 1995 to 2022, highlighting noteworthy patterns observed throughout the time series.

2. Materials and Methods

2.1. Study Area

The MABH has approximately six million inhabitants and an area of 9468 km^2 [25], across 34 municipalities, and serves as a prominent commercial, industrial, cultural, and tourist hub within the state of Minas Gerais. The area experiences significant heavy traffic and hosts an industrial complex with considerable potential for emitting air pollutants. In particular, the 2015 state emissions inventory for MABH [26] identified road traffic sources (dust resuspension + exhaust) and industrial sources as the main contributors to particulate matter, sulfur, and nitrogen oxide emissions. Biomass burning was identified as the third-largest contributor in the region.

For this study, data from 17 air quality monitoring stations (Figure 1) in MABH were evaluated from the period spanning 1995 to 2022. This study considered hourly concentration data of particulate matter for particles equal to 10 μm in diameter and smaller (PM_{10}) and particles equal to 2.5 μm in diameter and smaller ($PM_{2.5}$), nitrogen dioxide (NO_2), ozone (O_3), sulfur dioxide (SO_2), and carbon monoxide (CO) from 2009 to 2022, which were obtained from [13]. Data from the years 1995 to 2008 were acquired from the annual reports [27,28] when available. From 2008 onwards, all data were provided in.xls files by the local environmental agency (FEAM), except for $PM_{2.5}$, in which the monitoring only started in the early of 2015. The network covers six of the thirty-four municipalities composing the MABH, including Belo Horizonte (BH01, BH02, BH03, and BH04), Betim (BT01, BT02, BT03), Contagem, Ibirité, São José da Lapa, and Brumadinho (Figure 1). A flow chart of the method used in this work is shown in Figure 2.

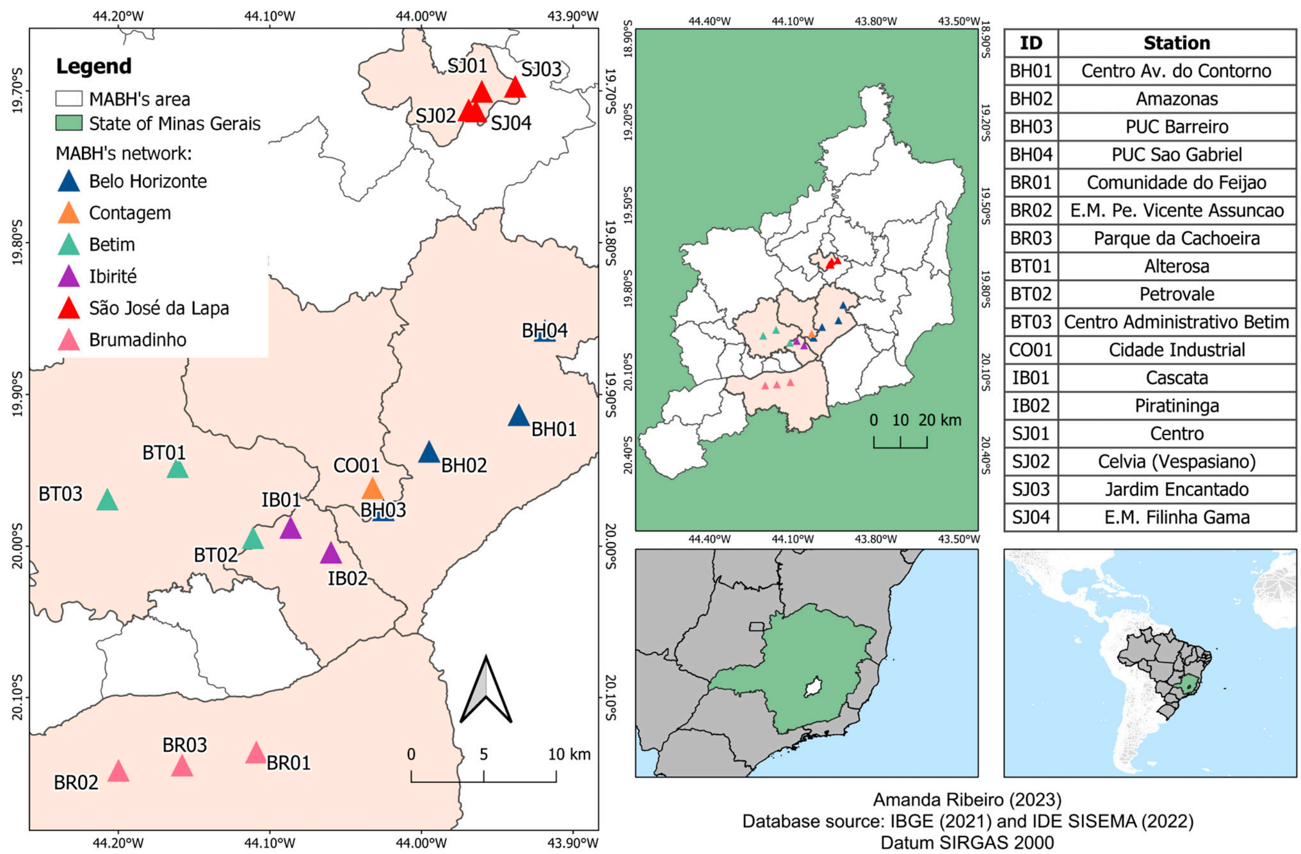


Figure 1. Locations of the MABHs air quality network.

2.2. Assessment of Air Quality Trends

The scope of this study encompassed an assessment of the MABH in relation to the WHO recommendations, taking into account both the interim targets (M1, M2, M3, and M4) and the final air quality guidelines (AQG). The interim targets are indicative stages that facilitate a gradual reduction of air pollution, ultimately leading to the attainment of air quality levels specified in the AQG. These AQG levels are established based on scientific evidence suggesting that adverse health effects either do not occur or are minimal below the specified concentration thresholds. These guidelines are presented in the latest issue of the WHO series of publications [11]. This paper focuses only on the long-term exposures (Table 1).

Table 1. Recommended AQG levels and interim targets for long-term exposures.

Pollutant	Averaging Time	Levels				
		Interim Targets				AQG
		1	2	3	4	
$PM_{2.5}$ µg/m^3	Annual [a]	35	25	15	10	5
PM_{10} µg/m^3	Annual [a]	70	50	30	20	15
O_3 µg/m^3	Peak season [b]	100	70	-	-	60
NO_2 µg/m^3	Annual [a]	40	30	20	-	10

[a] Approximately 99th percentile (i.e., 3–4 exceedance days per year). [b] Average of daily maximum 8 h mean O_3 concentration in the six consecutive months with the highest six-month running-average O_3 concentration. Reference: Adapted from OMS (2021) [11].

Furthermore, the air quality trends over the past 26 years were subjected to analysis using the Mann–Kendall test, which provides a statistical assessment of the probability of positive or negative tendencies (α = 0.05). In this analysis, two key indicators were considered: (i) if the *p*-value is below 0.05, it suggests a significant tendency; and (ii) if S is positive, so is the tendency, which means the concentrations are increasing over the years. The average representativeness of the data was assessed following the Brazilian Environmental Ministry (MMA) recommendations by the MMA's guide [29]. The MMA's guide is the national guide that establishes guidelines for air quality monitoring in Brazil. Air pollutant local emissions data from the official state inventory [26] were also analyzed and used as support for discussions. It is important to highlight that, despite MABH consisting of 34 municipalities, the official inventory includes data from only three municipalities (Belo Horizonte, Contagem, and Betim), which together account for approximately 66% of the MABH population [25]. Thus, there is a lack of representativeness among the other 31 municipalities that account for 34% of the total emissions.

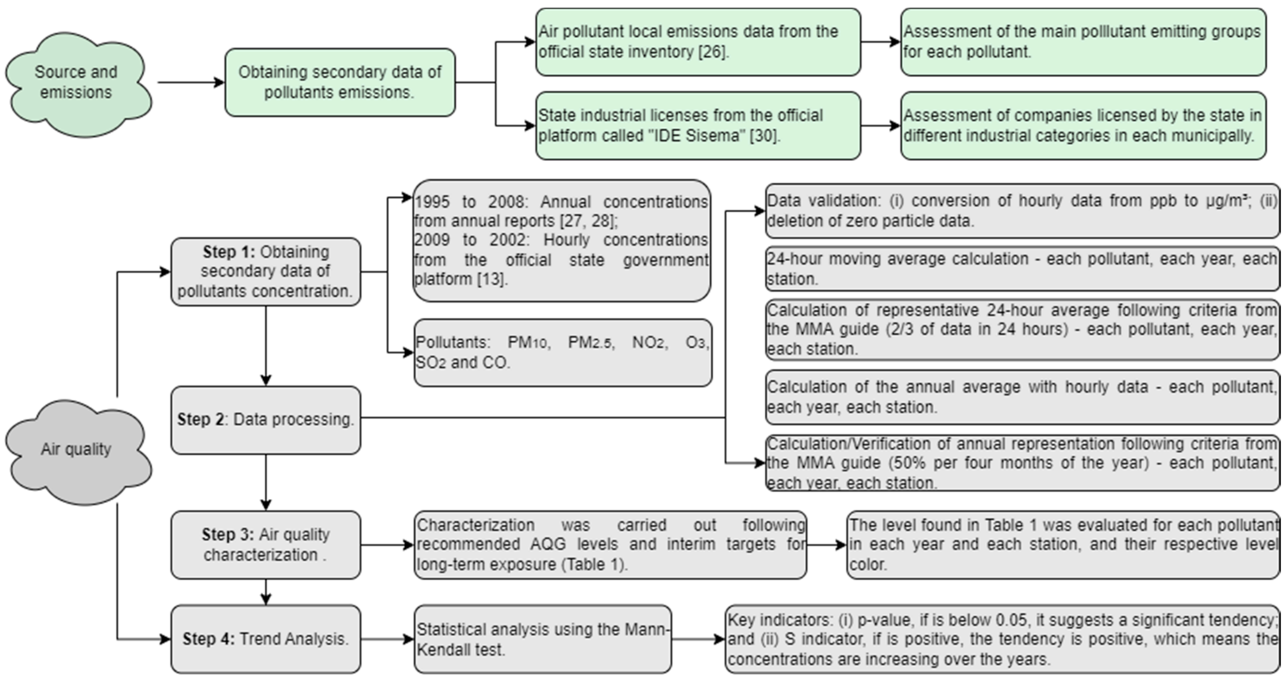


Figure 2. Flow chart of the methods used in the analysis of the trends and quality of the air pollution monitoring data.

3. Results and Discussion

3.1. Sources and Emissions

The current state of the MABH is characterized by emissions from road traffic and industrial sources. Figure 3 illustrates the total emissions of TSP (Total Suspended Particles), PM_{10}, $PM_{2.5}$, NOx (Nitrogen Oxides), SO_2 (Sulfur Dioxide), CO (Carbon Monoxide), and COV (Volatile Organic Compounds) based on the official inventory for the year 2015. The emissions are presented both for all source groups (Figure 3a) and specific industrial groups (Figure 3b). As the official inventory only includes data from three of all the 34 municipalities, it is important to highlight that consequently, the official inventory results might not fully represent all the cities within the metropolitan area, and there is a lack of official information available for the emissions of the remaining regions. Therefore, to gain a comprehensive understanding, an assessment was conducted by considering companies licensed by the state [30] under different industrial categories in municipalities with air quality monitoring stations within the state's network, as depicted in Figure 4.

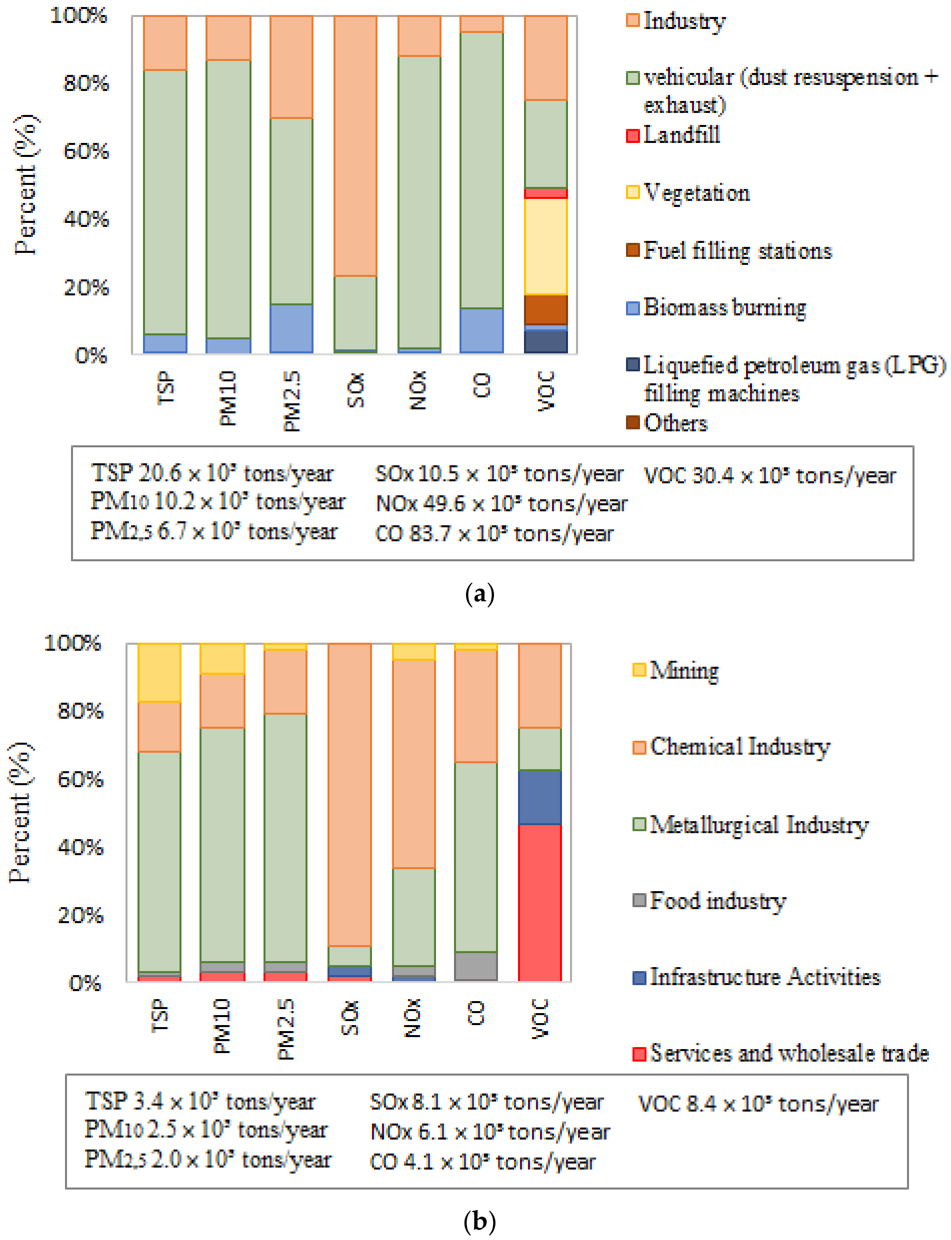


Figure 3. State official source inventory for the year 2015, in the MABH considering Belo Horizonte, Betim, and Contagem, in tons/year. (**a**) All sources groups. (**b**) Industrial groups.

Based on the inventory data, road traffic is a significant contributor to emissions, accounting for 78% of TSP emissions, 22% of SO_2 emissions, 86% of NO_2 emissions, 81% of CO emissions, and 26% of COV emissions [26]. Regarding the road traffic contributions to particulate emissions, the inventory combined dust resuspension with exhaust emissions as a single road traffic group. This approach presents a limitation as each component originates from different sources and necessitates distinct control mechanisms. Consequently, the official inventory fails to provide specific information about the percentage attributed to each type of road traffic emission in the MABH area, indicating a weakness in the availability of comprehensive information.

Santos et al. [24] also aimed to estimate traffic-related emissions in the MABH for the year 2015. They reported TSP emissions of approximately 2.4 × 10^3 tons/year from traffic sources, whereas the state inventory estimated much higher emissions at around 16 × 10^3 tons/year, nearly seven times greater than the former estimate. It is worth noting that the authors considered emissions from all 34 municipalities in MABH, while the official data only included three of them. Considering only Belo Horizonte, the state inventory estimated road traffic emissions of 11 × 10^3 tons/year for TSP, 27 × 10^3 tons/year for NOx, 1.7 × 10^3 tons/year for SOx, 52 × 10^3 tons/year for CO, and 6.4 × 10^3 tons/year for VOC. However, Santos et al. [24] estimated only 1 × 10^3 tons/year, 581 tons/year, 581 tons/year, 17 × 10^3 tons/year, and 4.7 × 10^3 tons/year, respectively.

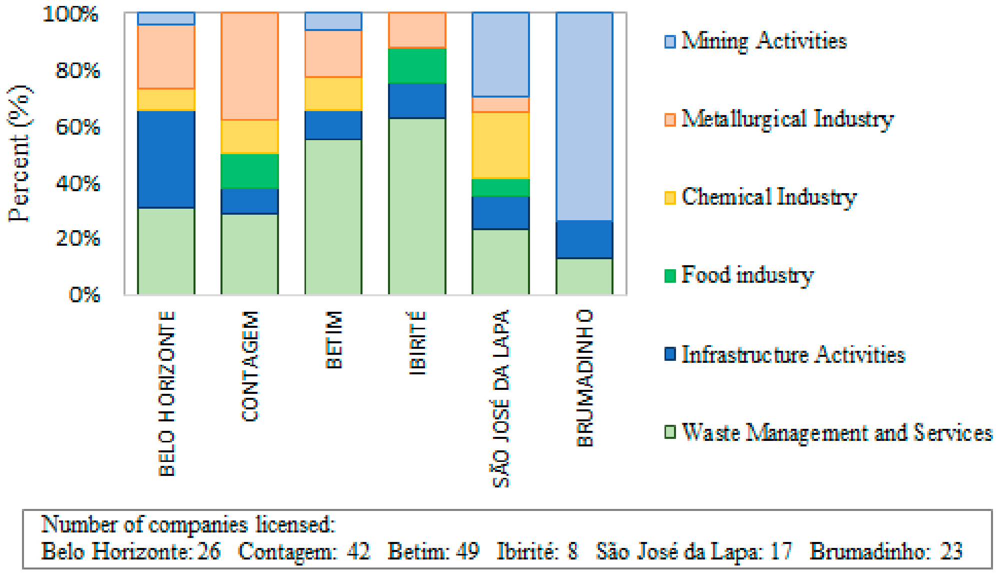


Figure 4. State licenses by industrial category for each city covered by the air quality network: Belo Horizonte; Betim; Contagem; Brumadinho; Ibirité; and São José da Lapa.

Particulate matter emissions from transportation in the Belo Horizonte region were found to exceed those of São Paulo [16], the most populous city in Brazil and the fourth most populous in the world, encompassing the largest metropolitan region in the country. This finding raises the possibility that the predominant contribution of road traffic emissions may be overestimated or, at the very least, sheds light on the inherent fragility of the state's emission estimate from road traffic circulation and its respective contribution percentage.

Regarding TSP, PM_{10}, $PM_{2.5}$, and CO emissions, the industrial and biomass burning emissions groups constitute the other two significant contributors. The industrial group's particulate and CO emissions are primarily associated with metallurgical industries, whereas SO_2 emissions are related to chemical plants. However, it is essential to acknowledge that these results do not fully represent all the cities within MABH. Therefore, to gain a comprehensive understanding, an assessment was conducted by considering companies licensed by the state [30] under different industrial categories in municipalities with air quality monitoring stations within the state's network, as depicted in Figure 4. The analysis reveals a variation in predominant emission typologies across different municipalities. Specifically, in Belo Horizonte, Contagem, Betim, and Ibirité, the metallurgical industry sector, waste management, and services, along with infrastructure activities, dominate the emissions profile. However, São José da Lapa and Brumadinho show a more prominent presence of mining activities, which significantly influence particulate matter emissions in these cities. São José da Lapa also has a notable presence in chemical industries, waste management, and services. However, the primary sector of production is mineral extraction and limestone processing, which are situated within the urban center of the city [31]. This difference between the categories of emission sources in the municipalities underscores the complexity of air quality management in the MABH due to the variability of emission sources across different municipalities, making it crucial to consider regional-specific factors when conducting emission assessments and air quality analyses.

Santos et al. [24] also estimated stationary emissions of MABH in 2015, showing similar results for all pollutants from industrial emissions when compared with the official inventory of MABH. However, more than five years have passed since these works, and an inventory review is required.

That way, the emission inventory in the MABH faces certain challenges. Nonetheless, the primary contributors to PM_{10} emissions in this region are road traffic, industrial, and biomass burning sources. The road traffic group, in particular, warrants attention, given the significant growth in the general fleet of vehicles, especially motorcycles and private vehicles, which surged by 76.6% between 2001 and 2009 [32], whereas MABHs population increased by 13.5%. It is important to note that heavy vehicles in the state of Minas Gerais, including the MABH region, were found to be the major contributors to emissions of particulate matter, including TSP, PM_{10}, and $PM_{2.5}$, in 2015, as highlighted by Santos et al. [24]. The emission breakdown for PM_{10} indicated that 48.8% originated from exhaust, 32.1% from tire and brake wear, and 19.1% from road wear. Consequently, it becomes evident that a significant portion of total PM_{10} emissions originates from non-exhaust sources [24], aligning with observations from studies conducted in other urban centers globally. The material composition of emitted particles is influenced by various factors, such as the type of engine, fuel, vehicle age, maintenance level, and the origin of deposited materials susceptible to resuspension [33–35]. An insightful study by Moura [36] analyzing the composition of PM_{10} in the MABH identified road traffic activities as a major emission source, alongside mining activities and soil dust resuspension. Regions like São José da Lapa and Brumadinho, characterized by substantial mining activities and waste management and services, contribute significantly to this pollutant's emission profile (Figure 4).

3.2. Air Quality Status and Tendency

Regarding the annual concentrations in the MABH, all pollutants exceeded the AQG levels (Figure 5). Notably, for NO_2, the concentrations surpassed the first interim target, M1 = 40 $\mu g \cdot m^{-3}$, showing a decreasing trend between 1995 and 2022. Nowadays, NO_2 concentrations vary between the M2 and M3 targets. The PM_{10} levels remained in the M2 phase most of the time, including in more recent years, whereas $PM_{2.5}$ showed concentrations at the M3 level since monitoring began. Furthermore, a drop in PM2.5 concentrations was observed in 2020 compared to other years. Consequently, for all pollutants with annual guidelines, the MABH failed to comply with the WHOs recommendations, highlighting the associated risk to the local population. This observation aligns with other studies demonstrating similar situations of pollutant concentrations exceeding recommended levels in some Brazilian and South American urban centers [7,10,16].

For O_3, SO_2, and CO, there are no specific annual levels recommended by the WHO. Nevertheless, a decrease in the average concentrations of O_3 and CO is noticeable in the MABH. There is no WHO 2021 recommended value for the annual average of SO_2; however, the results show that its concentrations did not present any decreasing tendency in relation to the current and first interim of the Brazilian air quality standard of 40 $\mu g/m^3$. Thus, while the annual concentrations of SO_2 have not decreased since 1995, they remain below the final standard of Brazilian legislation. Comparatively, in other Brazilian areas and South American cities [16], both CO and SO_2 have not emerged as significant problems in urban centers, primarily due to advancements in vehicle technology and fuels [37,38].

Considering the risks of these pollutants, it is important to cite the ELAPSE project. The ELAPSE project is a Europe-wide initiative spanning from mid-2016 to mid-2019. It delves into the effects of long-term exposure to low-level air pollutants like $PM_{2.5}$, Black Carbon, NO_2, and O_3. The project's focus lies in investigating the health impacts—both mortality and morbidity—stemming from these pollutants. It leverages detailed individual data from around 380,000 subjects in specific cohorts of the ESCAPE study, as well as data from seven extensive European administrative cohorts comprising over 35 million subjects [39]. It is important to highlight that the ELAPSE project found adverse health effects even for air pollutant concentrations below WHO-AQG (2021) levels, demonstrating that even for pollutants levels below WHO-AQG, there is still risk for adverse health effects.

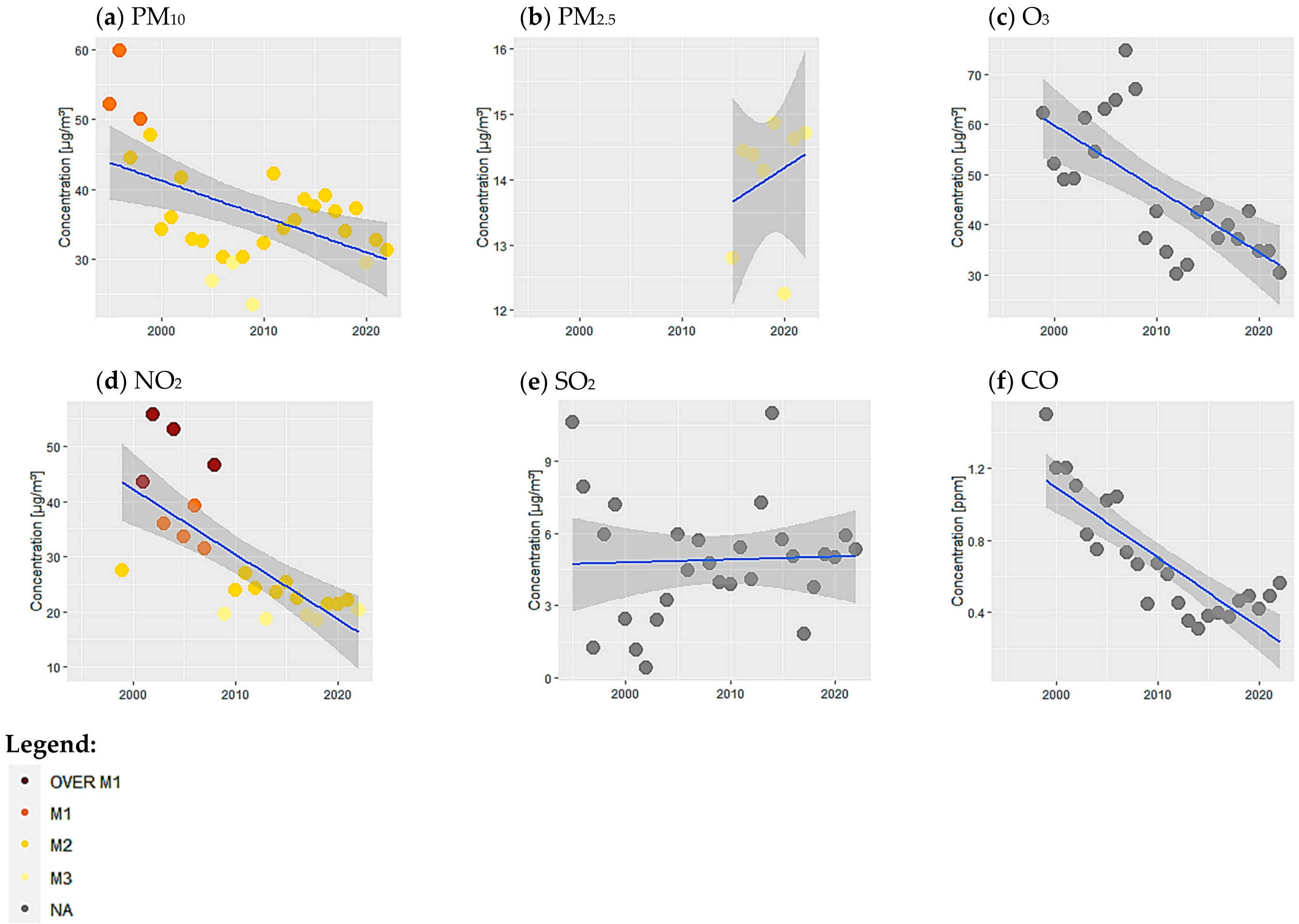


Figure 5. Evolution between 1995 and 2022 of the average values of pollutant concentrations measured at the MABH air quality monitoring for annual concentrations of (**a**) PM_{10}, (**b**) $PM_{2.5}$, (**c**) O_3, (**d**) NO_2, (**e**) SO_2, and (**f**) CO. Note: NA—Not Apply.

It is crucial to highlight that the Brazilian air quality standards are ruled by CONAMA Resolution No. 491/2018 [40], which established three interim standards and a final one, corresponding to the WHO recommendations at that time, the Global Update 2005 [41]. However, in 2021, a new edition of the WHO global air quality guidelines was published [11], offering updated recommendations for safeguarding public health from the adverse effects of air pollutants. Additionally, the fact that the final Brazilian standard is outdated, coupled with the country still adhering to the first interim standard with no migration deadline for the subsequent stages, underscores the nation's challenges in effectively monitoring and improving air pollutant concentrations throughout the Brazilian territory.

3.3. PM_{10} Averages and Trends

The analysis of the PM_{10} time series (Figure 6) reveals that annual concentrations consistently exceeded the established AQG levels for nearly all monitoring stations. Particularly concerning are three stations located in Contagem and Betim municipalities, which exhibit the most severe air quality degradation in the MABH. However, the stations of Contagem and Betim showed significant improvements in the air quality, as demonstrated by the Mann–Whitney test presented in Table 2. Despite this progress, it is crucial to acknowledge that PM_{10} concentrations in these regions still remain above the WHO guidelines. Throughout the entire monitoring period, the CT01, BT01, and BT03 monitoring stations registered average annual PM_{10} concentrations above 50 $\mu g/m^3$, with BT01 Station reaching levels exceeding 70 µg/m^3 (Figure 5). Comparatively, Peláez et al. [16] analyzed air quality and trends in major cities across South America, revealing that long-term PM_{10} concentrations in all studied cities exceeded WHO-AQG levels. Notably, Belo Horizonte reported higher annual averages of PM_{10} when compared to several Brazilian regions, including Vitória, São Paulo, Rio de Janeiro, and Salvador [16]. In contrast, Belo Horizonte displayed relatively lower PM_{10} concentrations compared to other prominent South American cities such as Bogotá, Medellín, Quito, Santiago, and Lima. Among the South American regions outside Brazil, only Buenos Aires recorded lower PM_{10} concentrations than Belo Horizonte. Thus, the findings underscore the persistent challenge of achieving and maintaining satisfactory air quality in the MABH, with PM_{10} concentrations posing significant health risks to the population. To address this issue effectively, it is essential to focus efforts on identifying the sources of PM_{10} pollution and implementing targeted control mechanisms. The complexity of PM_{10} pollution extends not only to other regions within Brazil [16,37,42,43] but also to various areas around the globe [16].

City	Station	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022	Tendency
Belo Horizonte	BH01	43	22	22	25	30	13	21	26	39	29	22	26	26	28	29	32	52	31		31	22	19	22	30	33	27	30	26	*
	BH02								17	18	17	14	17	17	15	15	12					31	27	32	22	21	17	24		*
	BH03																								27	31	26	27	27	*
	BH04																									27	21	26	27	*
Contagem	CO01	65	66	67	52	44	32	39	48	44	48	40	48									47	42	40	39	43	35	38		-
Betim	BT01	48	92		73	70	58	47	52	37	21	41	51	49	49	32	42	34	37	39	30	42	45	34	35	38	24	31	21	-
	BT02										41	31	41	41	38	23	32	36	31	34	34	34	29	29	26	25	17	20	18	-
	BT03											47	40	56	50	29	63	58	51	49	50	41	42	32	28	31	26	33	30	-
Ibirité	IB01									56		13	11	15	19	17	25	31	33	31	35	28	43	38	28	28	25	22	15	*
	IB02											18	20	12	23	18	25		25	24	51	23	41	32	27	39	27	28	26	+
São José da Lapa	SJ01																					68	68	60	63	70	59	71	62	*
	SJ02																					31	26	22	22	27	22	27	25	*
	SJ03																					43	39	48	50	52	42	34	43	*
	SJ04																					40	45	50	50	58	56	52	53	+
Brumadinho	BR01																					37	42	39	27	29	22	26		*
	BR02																									39	28	29		*
	BR03																									42	22	35		*

WHO TARGETS (2021):
- Over 70 µg/m³
- M1 50 to 70 µg/m³
- M2 30 to 50 µg/m³
- M3 20 to 30 µg/m³
- M4 15 to 20 µg/m³
- AQG Below 15 µg/m³
- Non-representative annual average (Criteria: 75% until 2008 and 50% per four-month period from 2009)

TENDENCY:
- + Tendency to increase
- - Tendency to decrease
- * Stability tendency

Figure 6. Average annual concentrations of PM_{10}, in µg/m^3, between 1995 and 2022, classified according to WHO air quality guidelines.

Table 2. Mann–Kendall test results between 1995 and 2022 for PM_{10}, $PM_{2.5}$, NO_2, and O_3.

City	Station	PM_{10} (1995–2022)			$PM_{2.5}$ (2015–2022)			NO_2 (1995–2022)			O_3 (1995–2022)		
		p-Value	S	Tendency	*p*-Value	S	Tendency	*p*-Value	S	Tendency	*p*-Value	S	Tendency
Belo Horizonte	BH01	0.25	56	None	0.80	3	None	0.00	−125	Negative	0.10	31	None
	BH02	0.13	35	None	0.55	5	None	1.00	0	None	0.39	−8	None
	BH03	1.00	0	None	0.81	2	None	1.00	0	None	0.46	−4	None
	BH04	0.73	2	None	1.00	0	None	0.73	−2	None	0.09	−6	None
Contagem	CO01	0.01	−79	Negative	0.00	21	Positive	0.39	−8	None	0.39	−8	None
Betim	BT01	0.00	−193	Negative	0.11	14	None	0.60	−6	None	1.00	−1	None
	BT02	0.00	−111	Negative	0.39	8	None	0.00	−83	Negative	0.00	−59	Negative
	BT03	0.01	−69	Negative	0.71	4	None	0.03	−59	Negative	0.36	−16	None
Ibirité	IB01	0.48	21	None	0.13	13	None	0.09	−49	None	0.13	−29	None
	IB02	0.01	66	Positive	0.62	5	None	0.00	−85	Negative	0.38	−17	None
São José da Lapa	SJ01	0.90	−2	None	0.54	6	None	-	-	-	-	-	-
	SJ02	0.71	−4	None	0.13	−13	None	-	-	-	-	-	-
	SJ03	0.90	−2	None	0.11	−14	None	-	-	-	-	-	-
	SJ04	0.04	18	Positive	0.27	10	None	-	-	-	-	-	-
Brumadinho	BR01	0.07	−13	None	1.00	−1	None	-	-	-	-	-	-
	BR02	1.00	−1	None	0.30	3	None	-	-	-	-	-	-
	BR03	1.00	−1	None	1.00	−1	None	-	-	-	-	-	-

Note: (i) if the *p*-value is below 0.05, it suggests a significant tendency; and (ii) if S is positive, it suggests a positive tendency.

Over the last eight years, the Air Quality Monitoring Station (AQMS) in São José da Lapa has experienced a concerning trend of high PM_{10} concentrations, surpassing the first target level for SJ01, SJ03, and SJ04 stations. This pattern of elevated PM_{10} levels is unique to São José da Lapa, as no other station exhibits a similar trend. The annual averages indicate a stable trend over the years, with no discernible signs of improvement or deterioration in PM_{10} concentrations. Figure 7, presenting a ranking of six stations with the highest averages from 2019 to 2021, further reinforces the severity of the situation. This specific period was chosen because all active stations were concurrently operational, providing a comprehensive overview. Within this context, the monitoring stations located in São José da Lapa (SJ01 and SJ04) recorded the highest and second-highest PM_{10} concentrations, respectively. These two stations are in close proximity to lime industrial plants in the region, suggesting a possible association between industrial activities and PM_{10} pollution. Additionally, Ireno [44] reported prevailing winds blowing from the industrial region toward the SJ01 station, indicating a high probability of particle transportation towards the station. Furthermore, both SJ01 and SJ04 stations are situated near major traffic roads, potentially contributing to the observed high PM_{10} levels. The third-highest average concentration was observed at SJ03, also located in São José da Lapa, further emphasizing the impact of local sources in the area. Notably, Contagem (CO01) and Brumadinho (BR02 and BR03) cities also recorded high average PM_{10} concentrations. CO01's ranking as the fourth-highest indicates the combined influence of industrial plants and road traffic emissions in the area. Meanwhile, the elevated PM_{10} concentrations in BR02 and BR03 stations in Brumadinho point to the presence of local sources related to mining industries in the vicinity.

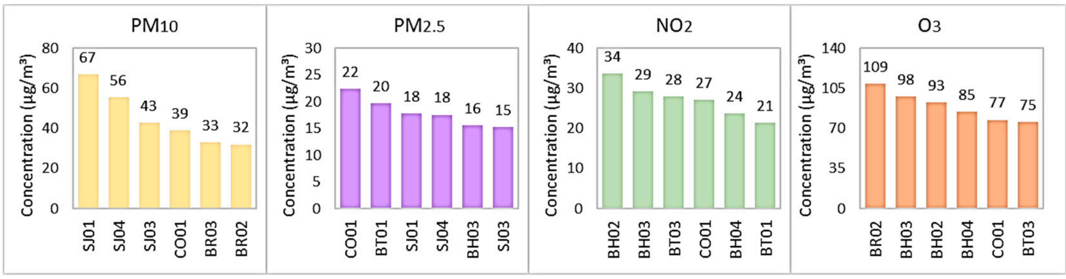


Figure 7. Ranking of the MABHs six stations with the highest last three-year averages (2019–2021).

The findings of this study indicate a substantial influence of industrial sources on PM_{10} pollution in the MABH. Notably, municipalities with the highest PM_{10} concentrations are significantly impacted by emissions from industrial activities, primarily stemming from the mining and steel industries. São José da Lapa, despite having a lower population density compared to other municipalities, exhibited PM_{10} concentrations well above those observed at other monitoring stations. This observation underscores the strong influence of industrial emissions on air quality in the region. The MABH, being the third-largest metropolitan area in Brazil, experiences a dual burden of pollution. Alongside pollution from urban sources, the region contends with an extensive industrial complex, further exacerbating air pollution challenges. As confirmed by Galvão et al. [45], in another industrialized region of southeastern Brazil, the mineral phases present in particulate matter are dependent on particle size. Hematite and quartz were found in significant amounts in the Settleable Particulate Matter (SPM) and in the Total Suspended Particles (TSP), whereas elemental carbon, sulfates, and halite were found in higher levels in the PM_{10} and $PM_{2.5}$ fractions [45]. This highlights the complexity of particulate matter composition and its potential implications for human health, as health risks are also influenced by PM size [18].

The burning of biomass emerges as one of the three largest groups of emitters contributing to PM_{10} pollution. A study conducted by [46] identified a concerning increase in fires over the years, with peaks in burned areas in Brazil often coinciding with extreme dry events. The extensive deforestation rates, particularly before 2005 and after 2019, have significantly impacted the rise in areas subject to burning [46]. Furthermore, the dry season, occurring between July and October, concentrates a staggering 83% of the country's burning activities [46]. The effects of biomass burning on more susceptible people, such as children and elders, were carried out in Brazil and revealed that an increase of 10 $\mu g \cdot m^{-3}$ of wildfire-related $PM_{2.5}$ was associated with a 1.65% (95% CI 1.51–1.80) increase in hospital admissions by all causes, besides an increase of 5.09% (4.73–5.44) of respiratory hospital admissions and a 1.10% (0.78–1.42) increase in cardiovascular hospital admissions over only 0 to 1 day after the exposure [47].

3.4. *$PM_{2.5}$ Averages and Trends*

$PM_{2.5}$ monitoring in the MABH area commenced in 2015 (Figure 8) and revealed concerning results. Throughout the monitoring period, the concentrations of $PM_{2.5}$ consistently exceeded the AQG values, with readings falling between the M4 and M2 targets. The trend analysis of the monitoring data displayed a stable pattern for most stations, suggesting no significant improvements in air quality over time. However, station CO01 stood out as an exception, showing a noticeable and significant increase in $PM_{2.5}$ concentrations over the last seven years. Despite this rise, the readings at CO01 remained within the M2 target throughout the entire time series. These findings indicate that Contagem, where station CO01 is located, experiences the most concerning air quality conditions regarding $PM_{2.5}$. The consistently high levels and the positive trend observed at CO01 emphasize the severity of the situation.

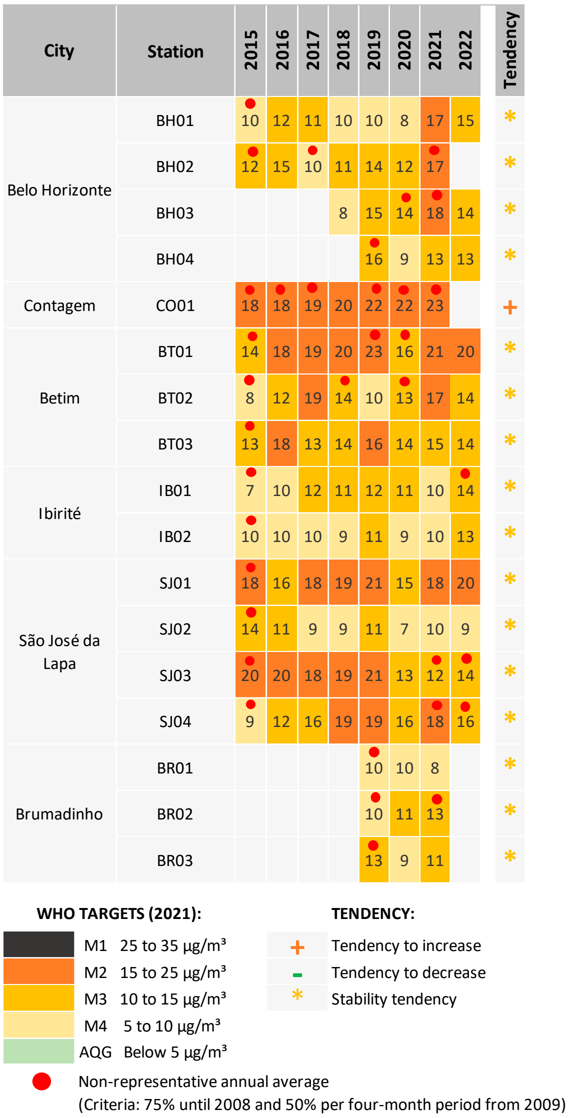


Figure 8. Average annual concentrations of $PM_{2.5}$, in $\mu g/m^3$, between 1995 and 2022, classified according to WHO air quality guidelines.

Figure 8 displays the $PM_{2.5}$ average concentrations for CO01 and BT01, which are the two stations with the highest readings. According to the official annual air quality report [48], BT01 is situated in an area with moderate road traffic and an industrial complex nearby. On the other hand, CO01 is located on the outskirts of the Industrial City of Contagem, close to heavily trafficked roads that connect the cities of Belo Horizonte, Contagem, and Betim. The stations SJ01, SJ04, and SJ03, located in the city of São José da Lapa, were also ranked among the worst cases in terms of $PM_{2.5}$ concentrations. Among the four São José da Lapa stations, only one did not fall under the worst concentrations for both fractions of particulates. This particular station is situated near an avenue in Vespasiano, at a greater distance from the region's lime industries. Another station with significant $PM_{2.5}$ levels is BH05, located in Belo Horizonte. In the vicinity of BH05, there are various pollution sources within a 1.5 km radius, including a railway line, a steel plant, and the industrial complex of Contagem.

Regarding $PM_{2.5}$ concentrations, the MABH area exhibited higher levels in regions with significant industrial influence. The stations CO01 and BT01, which recorded the two highest concentrations, were both affected by emissions from road traffic and industrial sources. According to the official state inventory [26], approximately 55% of $PM_{2.5}$

emissions in the MABH area originated from road traffic emissions, 30% from industrial activities, and 14% from biomass burning. Moreover, Galvão et al. [18] conducted a study that identified higher concentrations of particulate and gas-phase PAHs associated with fire outbreaks in the MABH region. The study pinpointed four potential sources in the area: gasoline-related (31.8%), diesel-related (25.1%), biomass burning (23.4%), and mixed sources (19.6%). Their findings also underscored the increased risks associated with exposure to $PM_{2.5}$-bound PAHs, particularly through ingestion and dermal contact, highlighting the urgency for mitigation and control measures to address $PM_{2.5}$ pollution in the region. Additionally, it is essential to consider the spatial context. Within a 40 km radius, the intensely urbanized area of the Belo Horizonte, Contagem, Betim, and Ibirité axes is located. As $PM_{2.5}$ represents the finest fraction of particulates, longer-range transport may also contribute to its presence in the region.

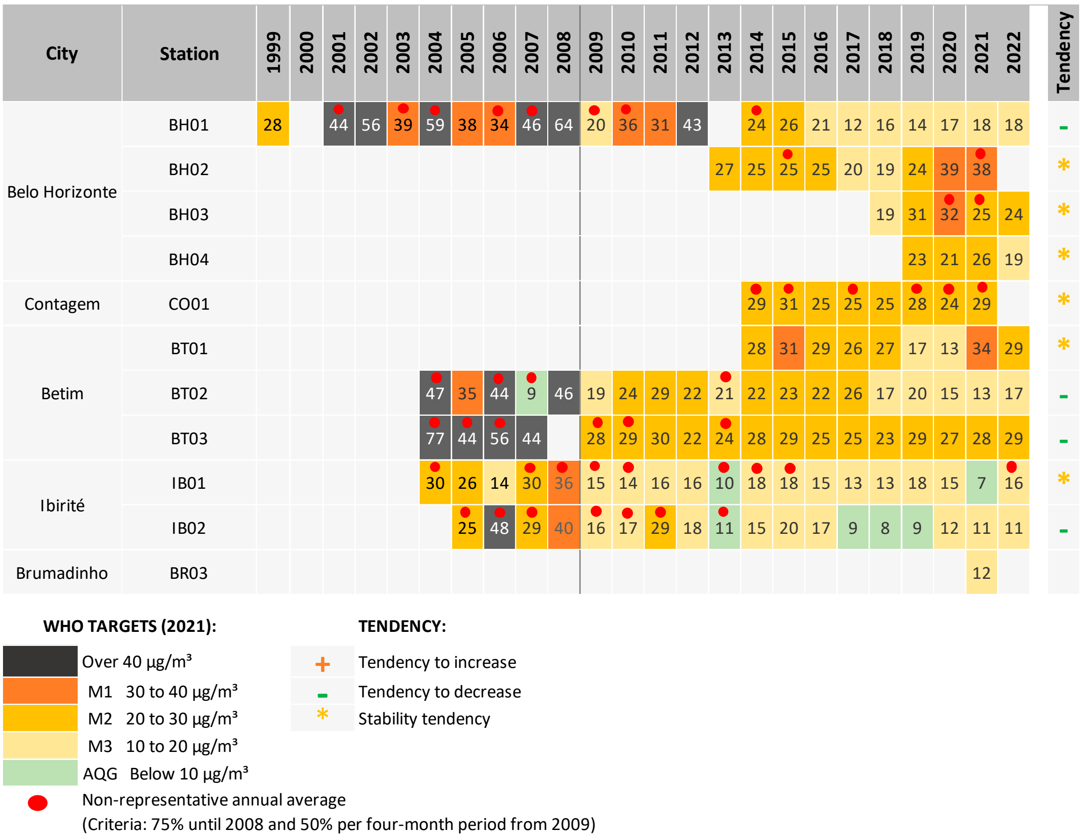
Regarding road traffic contributions, Santos et al. [24] found that heavy vehicles were the primary contributors to particulate emissions in the state of Minas Gerais. For $PM_{2.5}$, 63.9% of emissions originated from exhaust, while 22.5% came from tire wear and brakes, and 13.5% from road wear.

Peláez et al. [16] conducted a study showing that long-term concentrations of $PM_{2.5}$ in all the studied cities exceeded the WHO-AQG. In the case of Belo Horizonte, its annual $PM_{2.5}$ concentrations were higher than Vitória's but lower than São Paulo's. However, this difference decreased over the years between 2014 and 2017, with Belo Horizonte's annual average $PM_{2.5}$ concentrations increasing while São Paulo's decreased. In comparison to other cities in South America, Belo Horizonte exhibited lower concentrations of $PM_{2.5}$ than cities like Bogotá, Medellín, Quito, Santiago, and Lima.

3.5. *NO_2 Averages and Trends*

Figure 9 illustrates the distribution of NO_2 concentrations, revealing that only Ibirité stations (IB01 and IB02) recorded annual averages below the Air Quality Guideline—AQG. However, the cities of Belo Horizonte (BH02, BH03, and BH04), Contagem (CO01), and Betim (BT01 and BT03) experienced the highest current NO_2 concentrations, falling within the range of the M1 and M2 interim targets. The official inventory [26] highlights that NO_2 emissions primarily originate from two sources: 86% from the road traffic group and 12% from the industrial sector. Within the industrial sector, 61% of NO_2 emissions come from chemical industries, while 29% stem from metallurgical industries. An examination of NOx emissions related to the fleet profile and fuel usage in Minas Gerais in 2015 [24] revealed that 68% of NOx emissions were attributed to trucks, with an additional 23% coming from buses. Remarkably, 94% of NOx emissions from vehicle exhaust were associated with diesel-powered vehicles.

Over the last three years, the monitoring data revealed that BH02, BH03, and BT03 were the stations with the most concerning air quality, as they consistently remained between the M1 and M2 interim targets for NO2 concentrations. Figure 7 further supported this finding, confirming that the municipalities of Belo Horizonte (BH02, BH03, and BH04), Contagem (CO01), and Betim (BT03) experienced the highest NO_2 levels. According to Peláez et al. [16], long-term concentrations of NO_2 in all the studied cities surpassed the WHO-AQG. However, it is noteworthy that Belo Horizonte exhibited relatively lower concentrations compared to other South American cities, despite exceeding the WHO-AQG. The results showed an improvement in air quality in locations where monitoring began before 2009. Stations BH01, BT02, BT03, and IB02, situated in Belo Horizonte, Betim, and Ibirité, respectively, experienced a significant decrease in NO_2 concentrations. However, other monitoring stations did not exhibit a clear trend regarding annual concentrations.



City	Station	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022	Tendency
Belo Horizonte	BH01	28		44	56	39	59	38	34	46	64	20	36	31	43		24	26	21	12	16	14	17	18	18	-
	BH02															27	25	25	25	20	19	24	39	38		*
	BH03																				19	31	32	25	24	*
	BH04																					23	21	26	19	*
Contagem	CO01																29	31	25	25	25	28	24	29		*
Betim	BT01																28	31	29	26	27	17	13	34	29	*
	BT02						47	35	44	9	46	19	24	29	22	21	22	23	22	26	17	20	15	13	17	-
	BT03						77	44	56	44		28	29	30	22	24	28	29	25	25	23	29	27	28	29	-
Ibirité	IB01						30	26	14	30	36	15	14	16	16	10	18	18	15	13	13	18	15	7	16	*
	IB02							25	48	29	40	16	17	29	18	11	15	20	17	9	8	9	12	11	11	-
Brumadinho	BR03																							12		

Figure 9. Average annual concentrations of NO_2, in µg/m^3, between 1995 and 2022, classified according to WHO air quality guidelines.

3.6. O_3 *Averages and Trends*

Regarding O_3, Figure 10 illustrates the evolution of maximum moving averages of 6 months, calculated from the maximum daily values of representative 8 h moving averages (with a 75% representation for 8 h), starting in June 2009. the WHO guidelines, there were only a few instances when ozone concentrations remained below the AQG level, as observed at monitoring stations BH01, IB01, and IB02. However, most of the time, ozone concentrations were found between the M1 and M2 interim targets. On specific occasions, particularly in 2019 and 2021 for SJ01 and in 1996 and 1998 for BT01, concentrations exceeded 70 µg/m^3, surpassing target levels. Throughout the time series, BH01 consistently displayed the best air quality among all the monitoring stations. Moreover, all the stations did not show any clear trend of improvement in air quality, except for BT02. The Mann–Whitney test indicated a negative trend for BT02, which could be attributed to the unusually high concentration in the first year of data collection in 2009, with an average exceeding 100 µg/m^3.

City	Station	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022	Tendency
Belo Horizonte	BH01	49	55	58	57		66	68	53	65	64	73	70	64	54	+
	BH02					88	83	99	109	78	67	93				*
	BH03										100	106	108	80	72	*
	BH04											91	83	80	74	*
Contagem	CO01						97	135	97	85	92	93	98	42		*
Betim	BT01	62	81	83	68	60	65	76	74	82	82	65	71	72	63	*
	BT02	103	88	95	78	75	79	86	86	82	81	76	75	74	68	-
	BT03		76	80	74	66	77	85	77	64	65	80	80	66	62	*
Ibirité	IB01	72	96	93	53	63	76	115	80	71	57	51	60	65	63	*
	IB02	73	80	86	71	58	78	84	84	70	65	65	58	76	77	*
Brumadinho	BR02													109		

WHO TARGETS (2021):

- Over 100 µg/m³
- M1 70 to 100 µg/m³
- M2 60 to 70 µg/m³
- AQG Below 60 µg/m³

TENDENCY:

- + Tendency to increase
- - Tendency to decrease
- * Stability tendency

Figure 10. The annual maximum peak season concentrations of O_3, in µg/m^3, between 1995 and 2022 were classified according to WHO air quality guidelines.

Based on the last three years (Figure 10), the municipalities of Brumadinho (BR02), Belo Horizonte (BH03, BH02, and BH04), Contagem (CO01), and Betim (BT03) were identified as the areas with the worst Ozone (O_3) pollution. Notably, Brumadinho's station (BR02), which began monitoring in 2021, recorded the highest ozone air pollution within the MABH network, with a value of 109 µg/m^3. These findings highlight the necessity for a comprehensive investigation into the factors driving the increase in ozone concentrations in this area. Aside from BR02, other stations ranking high for O_3 pollution also feature prominently in the air pollution rankings for Nitrogen Dioxide (NO_2), corroborating a potential correlation between high O_3 and NO_2 levels in these locations, warranting further scrutiny.

Figure 11 provides a visual representation of the historical evolution of hourly ozone concentrations in the MABHs air quality-monitoring network from 1999 to 2022, offering valuable insights into the long-term trends in ozone levels. The results from 1999 to 2022 indicate a similarity in the curves between Belo Horizonte and the entire MABH network, suggesting that O_3 pollution in Belo Horizonte might influence the trend of the metropolitan region. However, analyzing the period since 2009 (panel B, Figure 11), it becomes evident that although the time series showed an increase since 1999, a reversal occurred from 2009 onwards, indicating a slight decrease. Given that O_3 forms through the oxidation of atmospheric pollutants like volatile organic compounds (VOC) and NOx, it is crucial to investigate the formation processes and ideal conditions for reactivity. Understanding this is vital to finding effective ways to reduce O_3 levels in the MABH. The increase in O_3 concentrations, despite the presumed reduction in NO_2, highlights the significance of conducting a chemical speciation study and examining the potential influence of VOC on ozone formation, as demonstrated by Alvim et al. [49] in the São Paulo region (SP/Brazil) and Galvão et al. [50] in Vitória (ES/Brazil).

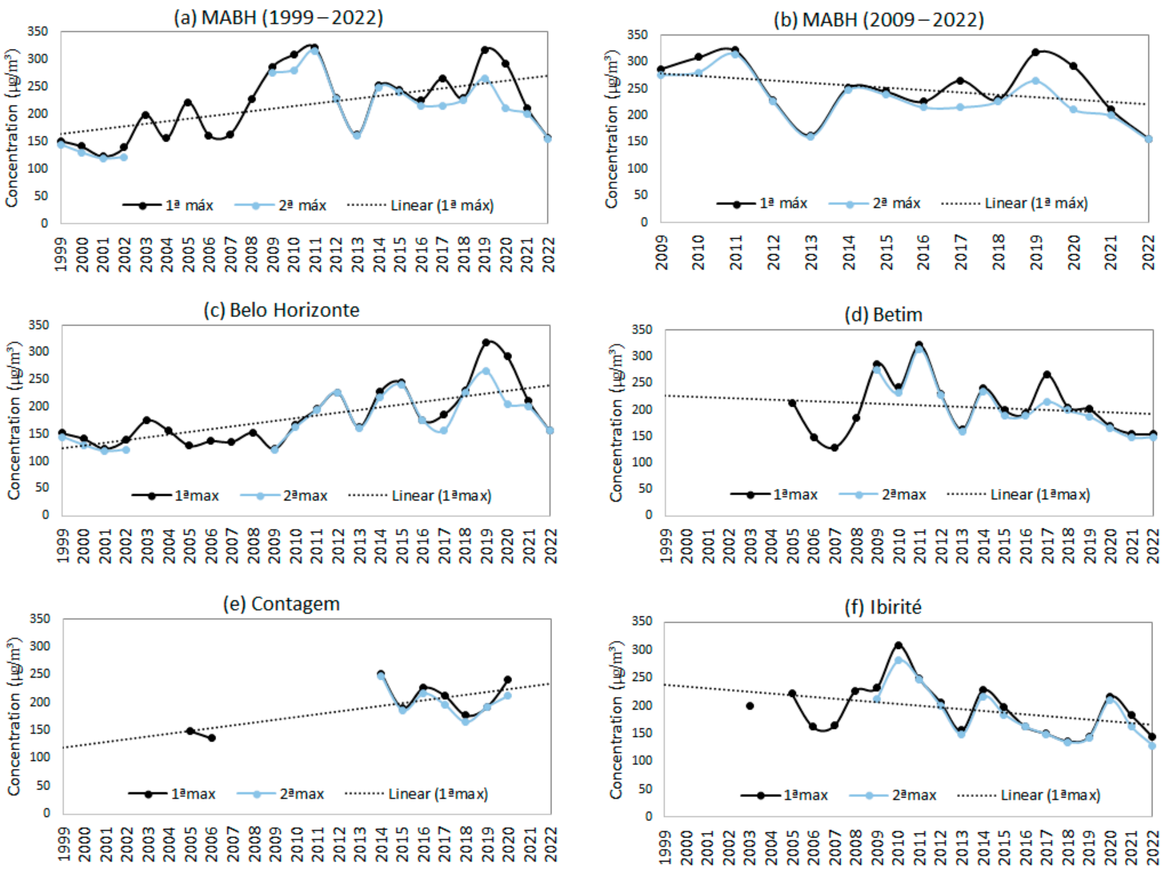


Figure 11. First and second maximum concentrations of hourly ozone at the MABHs air quality-monitoring network, from 1999 to 2022.

According to the local emissions inventory [26], the significant contributors to VOC emissions in 2015 were biomass burning (vegetation), road traffic, and industries, particularly service and trade groups, wholesalers, and chemical industries, as well as fuel stations, liquefied petroleum gas filling plants, and landfills. However, it is important to note the possibility of overestimating VOC emissions from vegetation in this inventory. Galvão et al. [18] identified potential emission sources of PAHs in Belo Horizonte, including fine fractions of particulates and VOCs, associated with road traffic sources (gasoline and diesel), biomass burning, and mixed sources (industries and road traffic emissions related to gasoline and natural gas combustion for industrial and domestic use). Regarding particulate emissions, the local emission inventory of 2015 [26] also highlighted biomass burning as one of the main contributors, even for smaller fractions, in the region.

Peláez et al. [16] conducted a short exposure analysis and revealed that among South American cities, Brazilian cities such as Belo Horizonte, São Paulo, and Rio de Janeiro exhibited the worst O_3 concentrations.

4. Conclusions

This study faces a shortage of air quality information and research focused on the MABH, which stands as the third most populous metropolitan region in Brazil. In this study, our primary objective was to comprehensively investigate the air quality characterization and trend of air pollutant concentrations within MABHs automatic network, spanning the period from 1995 to 2022.

In general, results showed that pollutants like NO_2, PM_{10}, and $PM_{2.5}$ consistently exceeded WHO-AQG, posing risks to the local population. There were decreasing trends in NO_2 concentrations and variations in PM levels, with a noticeable drop in $PM_{2.5}$ concentrations observed in 2020. O_3, CO, and SO_2 did not breach WHO-AQG, and there were decreases in average concentrations for O_3 and CO, whereas SO_2 levels remained below the final standard in Brazilian legislation, despite not meeting WHO recommendations. This aligns with similar situations in other Brazilian and South American urban centers, highlighting the widespread challenge of air quality management in the region. Therefore, collaborative efforts involving rigorous monitoring, comprehensive research, and effective interventions are needed to ensure improved air quality and safeguard public health.

The results highlight persistent issues with high PM_{10} concentrations in the MABH, particularly in stations located in Contagem, Betim, and São José da Lapa. Although some stations in Contagem and Betim show improvements in air quality, they still exceed WHO guidelines. São José da Lapa stands out for consistently high PM_{10} levels over eight years, potentially linked to industrial and traffic-related sources. The proximity of monitoring stations to industrial sites and major roads, combined with prevailing wind patterns, suggests a strong association between these factors and heightened PM_{10} pollution. Additionally, PM_{10} levels in Brumadinho indicate mining-related sources contributing to elevated concentrations. These findings underscore the complex challenge of addressing PM_{10} pollution in the MABH and emphasize the need for targeted strategies to identify and control pollution sources for better air quality.

The $PM_{2.5}$ monitoring from 2015 onwards consistently showed levels surpassing AQGs, notably falling between the M4 and M2 targets. While most locations displayed stable patterns, Contagem notably stood out for a significant and continuous increase in $PM_{2.5}$ concentrations over seven years, remaining within the M2 target. Contagem and Betim (BT01) stations exhibited the highest $PM_{2.5}$ levels, attributed to traffic and industrial emissions. São José da Lapa's stations also ranked poorly for $PM_{2.5}$ concentrations, mainly linked to industrial activities nearby. These findings align with reports indicating a majority of $PM_{2.5}$ emissions originate mainly from road traffic, industries, and biomass burning. The results highlighted the urgent need for mitigation strategies. Additionally, the spatial context of the intensely urbanized Belo Horizonte-Contagem-Betim-Ibirité axis suggests potential long-range transport contributing to $PM_{2.5}$ pollution in the region.

The analysis of ozone (O_3) pollution in the MABH also highlights significant trends and concerns. It shows a historical increase in ozone concentrations until 2009, followed by a slight decrease thereafter. Monitoring stations showed variations in air quality, with some stations in Belo Horizonte consistently displaying better conditions, whereas newer stations in Brumadinho record alarmingly high ozone levels. There is a notable correlation between areas with high O_3 pollution and elevated NO_2 levels, indicating a potential link between these pollutants. This emphasizes the urgency of investigating the factors contributing to increased O_3 concentrations, particularly in regions like Brumadinho, and understanding the role of pollutants like VOCs and NO_x in ozone formation. The need for a chemical speciation study is emphasized to devise effective strategies for mitigating ozone pollution in the MABH region.

Lastly, this study sheds light on the existing gaps in understanding and addressing air pollution issues in the MABH. By revealing the challenges and providing valuable insights into the sources and trends of air pollutants, we hope to foster greater awareness and drive effective actions toward ensuring healthier air quality for the inhabitants of this densely populated region. Further research and a concerted effort by relevant stakeholders are essential to mitigate the adverse impacts of air pollution and protect public health in MABH.

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