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Analysis of the particulate matter long term emissions in Romania by sectors of activities

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Abstract
Particulate matter (PM) in the atmosphere is highly relevant due to its far-reaching implications. These tiny solid or liquid particles affect human health, causing respiratory and cardiovascular issues, and even premature death. PM also plays a role in climate change, as some particles contribute to warming, while others have cooling effects, impacting Earth's energy balance. Additionally, PM influences visibility, transportation safety, and ecosystems by depositing on land and water surfaces, altering soil and water quality. Recognizing and monitoring the emissions and sources of PMs by 4 subclasses, from PM2.5 to PM10, but also as TSP (total suspended particles) and individually as BC – black carbon is vital for effective air quality management and mitigating its multifaceted impacts on our environment and well-being. This paper presents an analysis of PMs emissions divided into the 4 classes presented above, for Romania, between 1990-2021, as given by the national report made according to the National Emission reduction Commitments Directive (NECD), which aims to reduce emissions of the main air pollutants. The emission data are presented by national emission sectors, from different industries to different transport types, and also from agricultural activities, and waste management. Relevant averages and trends are quantified and analysed as a reference for real-time data at city level, such that to assess the local pollution level and data accuracy as compared to the national reports.

Keywords: pollution; particulate matter; emission sources, Romania.

1. INTRODUCTION

Particulate matter (PM) refers to tiny particles or droplets that stay aloft, given the equilibrium between the gravitational and the Archimedean forces. These particles can have various sizes, compositions, and origins, and they significantly influence air quality and climate, human health, and the overall well-being of the environment. PMs are significantly divided into 2 main classes: fine, PM2.5 (size less than 2.5 microns) and coarse, PM10 (size between 2.5 and 10 microns), since their size is essential with respect to most of their effects. For example, larger PMs contribute to reduced visibility, while small particles penetrate deeply into the human body, and induce not only respiratory problems, but also affect blood circulation and even the digestive apparatus. Fine PMs include metals, organic compounds and combustion particles, as black carbon, while coarse PMs can be dust, mould, pollen etc. [1-5].

Particulate matter can have both cooling and warming effects on climate. While some particles reflect sunlight, others absorb it, contributing to warming (i.e. they have a greenhouse effect). The hygroscopic PMs can absorb moisture from the surrounding air. This hygroscopic nature influences the water content of the particles and, in turn, affects the ambient humidity and cloud formation processes. Thus, PMs influence the water status in the atmosphere. Particulate matter serves as cloud condensation nuclei by providing surfaces on which water vapour can condense or even desublimate to form tiny droplets or ice crystals. Without PMs, water vapour would need supersaturation levels to condense into droplets. PMs can also serve as ice nucleation sites, promoting the freezing of water droplets into ice crystals, which is significant in the upper levels of the atmosphere where temperatures are very low. Thus, cloud formation at any altitude, is strongly influenced by the atmospheric PM content. And since clouds reflect and absorb radiation, aerosols have an indirect effect on weather and climate. More than that, larger PM particles may enhance the coalescence of droplets, leading to the formation of larger precipitation particles that fall to the ground, while smaller particles can contribute to the persistence of smaller cloud droplets. Through precipitation, PMs are removed from the atmosphere, but, instead, contaminate the soil and water, affecting the aquatic life, but also plant productivity not only indirectly, through soil pollution, but also directly, by depositing on leaves and diminishing the photosynthesis process [2-8].

A specific class of PMs is considered, given its specificity as effects: black carbon (BC), which consists of carbon particles (most often, less than 2.5 microns) that are released into the atmosphere through incomplete combustion of fossil (i.e. carbon-based) fuels. Along with the typical consequences of PMs of pollution, visibility decrease, and being harmful to human health, the dark colour of BC makes it absorb sunlight, contributing to the overall heat retention in the atmosphere.

The term 'total suspended particles' (TSP) denominates all solid and liquid particles and even their combination within the air that are small enough to be suspended rather than settled. TSP includes particles of various sizes, ranging from coarse particles to fine particles, namely all PMs. Recently, some researchers consider as TSP only those PMs bigger than 10 microns, thus bigger than PM10, namely with sizes up to 100 - 120 microns [1, 9-14].

As most pollutants, PMs can be transported over long distances from their source, and thus, contribute to transboundary air pollution. Wildfires, volcanic eruptions, and dust storms represent natural sources of PMs, while the anthropogenic sources include industry, transportation, construction, household activities and waste management. Different types of transportation modes induce different PM emissions, especially road transportation, from exhaust and paved roads, but also from tyre and brake wear [15-17], but also different industries, from large scale ones, as steel plants, to small scale ones, such as smelters and spray paint factories [18]. There is already some time since thorough studies revealed that PM2.5, PM10 and BC resulting from traffic within even 300m of roadways are associated with increased prevalence of asthma or other respiratory symptoms [5, 7, 10, 19-21]. Agricultural activities are also relevant sources of particulate matter emissions. Thus, land preparation, crop harvesting and grain handling, agrochemical and manure application as fertilizers, and also the animal feeding operations determine emissions of PM2.5, PM10 and TSP, while pollen emissions are included within the TSP class [11, 13, 14, 22].

Many countries have established air quality standards and regulations to limit the concentration of particulate matter in the atmosphere. Monitoring and controlling PM levels are crucial for public health and environmental protection. The national monitoring networks have specific positions for their groups of sensors, which were established according to the physical-geographical an meteorological characteristics of the areas to be monitored. At the European Union level, the component countries have the obligation to report yearly the emissions of different types of pollutants from different types of human activities. Currently, there are already long databases at national levels, including for Romania. Such databases allow for assessing which activities give the most important emissions of pollutants, such that measures could be taken to reduce them, especially directly at source and also, to assess if the imposed regulations and the measures already taken are efficient, fact which can be seen as pollutant reduction. This paper represents a study of the PM emissions in Romania from anthropogenic sources for 32 years.

2. DATA TYPES AND SOURCES

The PM data were taken for Romania, between 1990 and 2021, from ANNEX 1: National sector emissions: Main pollutants, particulate matter, heavy metals and persistent organic pollutants from the European Environment Agency database [23]. The monitored parameters were PM2.5, PM10, TSP and BC. The data are expressed as annual mass emissions, in kilotons (kt). The sectors of human activities, considered for emissions, were [23]:

1. public electricity and heat production
2. industry (44 types)
3. transportation – as all types (14 types) and separate: road transport, aviation, railways, water transportation
4. agriculture
5. wastes
6. fugitive (i.e. undirected emissions, released into the surrounding air through windows, doors and other openings, ventilation systems or openings, which do not normally fall into the category of directed sources of pollution)

Sums were computed for the different subsectors within the same sector of human activity.

Thus, there are 44 ***industrial*** subsectors, excluding the public electricity and heat production, among which: petroleum refining; manufacture of solid fuels and other energy industries; stationary combustion in manufacturing industries and construction from: iron and steel, non-ferrous metals, chemicals, pulp, paper and print, food processing, beverages and tobacco etc.; mobile combustion in manufacturing industries and construction; cement, lime, and glass production, respectively (3 types); construction and demolition; production of different minerals, chemicals and metals, their storage, handling and transport; wood processing; production and consumption of POPs (persistent organic pollutants) and heavy metals.

For ***Agriculture***, 2 classes of emission sources were considered, with a total of 26 subtypes:

- emissions from livestock concern emissions from manure management from 13 types of livestock: dairy cattle, non-dairy cattle, sheep, swine, buffalo, goats, horses, mules and assess, laying hens, broilers, turkeys, other poultry and other animals.
- emissions from crops include: 1) organic n-fertilizers; 2) animal manure applied to soils; sewage sludge applied to soils; 3) other organic fertilizers applied to soils (including compost); 4) uri and dung deposited by grazing animals; 5) crop residues applied to soils; 6) indirect emissions from managed soils; 7) farm-level agricultural operations including storage, handling and transport of agricultural products; 8) off-farm storage, handling and transport of bulk agricultural products; 9) cultivated crops; 10) use of pesticides; 11) field burning of agricultural residues; 12) agriculture other.

There are 14 types of emissions sources from ***transportation***, divided into:

- ***railways***, with no subsectors;
- ***aviation***, which comprises: i) international aviation LTO (civil); ii) Domestic aviation LTO (civil); iii) International aviation cruise (civil); iv) Domestic aviation cruise (civil);
- ***road transportation*** which includes: 1) passenger cars; 2) light duty vehicles; 3) heavy duty vehicles and buses; 4) mopeds & motorcycles; 5) gasoline evaporation; 6) automobile tyre and brake wear and 7) automobile road abrasion;
- ***water transportation***, which includes: 1) international inland waterways and 2) international navigation (shipping).

The ***wastes subsectors*** comprise 15 types of emission sources: biological treatment of waste through: solid waste disposal on land, composting, and through aerobic digestion at biogas facilities, respectively (3 types); municipal, industrial, hazardous, and clinical waste incineration, respectively (4 types); sewage sludge incineration; cremation; other waste incineration; open burning of waste; domestic wastewater handling; industrial wastewater handling; other wastewater handling; other waste.

Finally, there are 9 subsectors for ***fugitive emissions:*** fugitive emission from solid fuels: coal mining and handling; fugitive emission from solid fuels: solid fuel transformation; other fugitive emissions from solid fuels; fugitive emissions oil: exploration, production, transport; fugitive emissions oil: refining and storage; distribution of oil products; fugitive emissions from natural gas; venting and flaring; other fugitive emissions from energy production.

3. RESULTS AND DISCUSSION

Fig. 1 presents the annual emissions of four different classes of PMs in Romania, between 1990 – 2021, namely PM2.5, PM10, TSP and BC, respectively. It can be noticed that each type of investigated particulate matter has an increasing trend for Romania, between 1990 and 2021, with some local peaks, especially in 1997 and 2008. After 2012, BC presents a relative stagnation in emissions, with a slight increase from 2020.

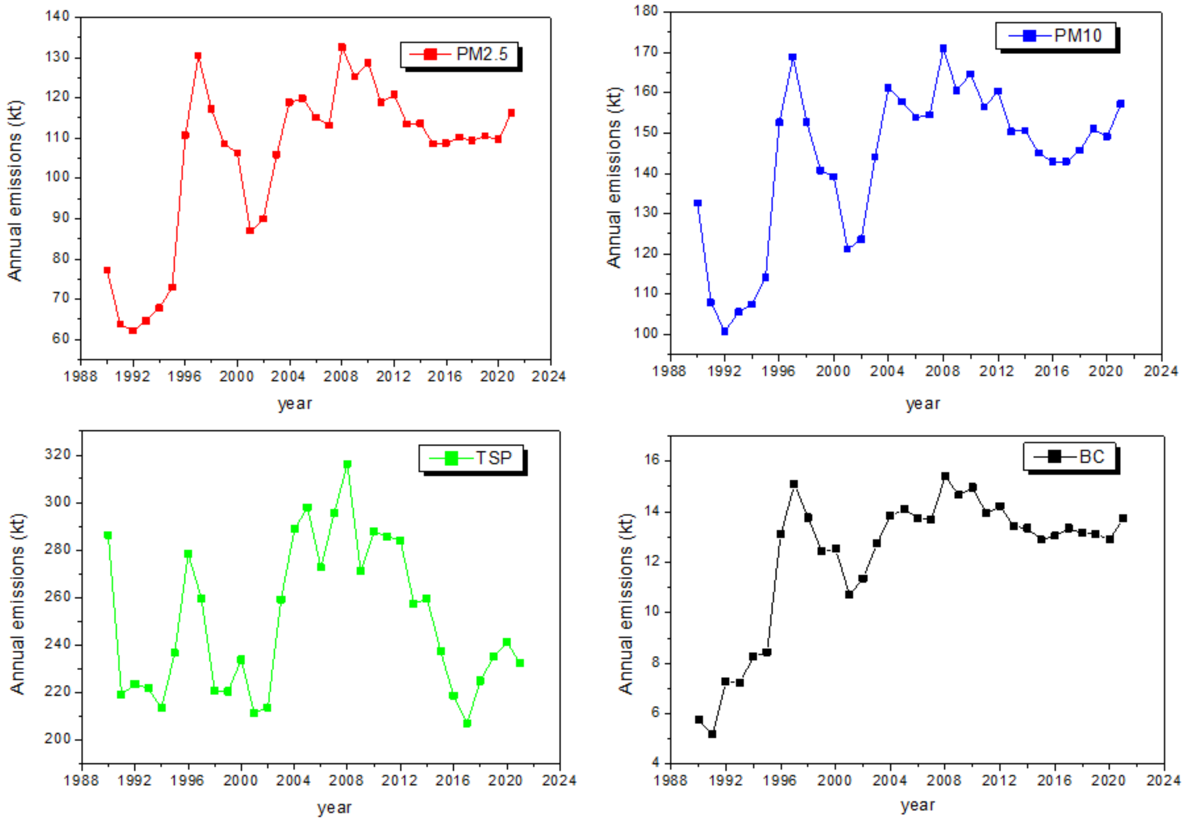

Fig. 1. Annual emissions of different classes of PMs in Romania, between 1990 – 2021

Fig. 2 presents a comparison between the annual emissions of PM2.5 and PM10 in Romania, within 32 consecutive years. It can be seen that there were higher emissions for PM2.5 than for PM10, while their trends are similar.

Figs. 3 and 4 present the evolution of the annual emissions of PM2.5, PM10, TSP and BC in Romania, for the 6 sectors of human activities described in the previous section of this paper. For PM2.5, one can notice that, in average, the highest emissions are from industry, while the lowest are from wastes. Also, there is an accentuated decrease in emissions from public energy and heat production after 2011, while there are steady emissions from transportation from 2003 till now. For PM10, half of the time corresponds to highest emissions from public energy and heat production and half from industry, while lowest emissions are from wastes. Also, there is an accentuated emission decrease from public energy and heat production after 2011, while the emissions from agriculture increase from 2016.

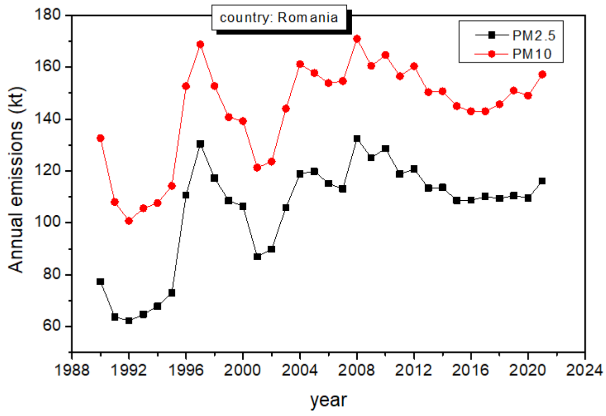

Fig. 2. Comparison between the annual emissions of PM2.5 and PM10 in Romania, between 1990 – 2021

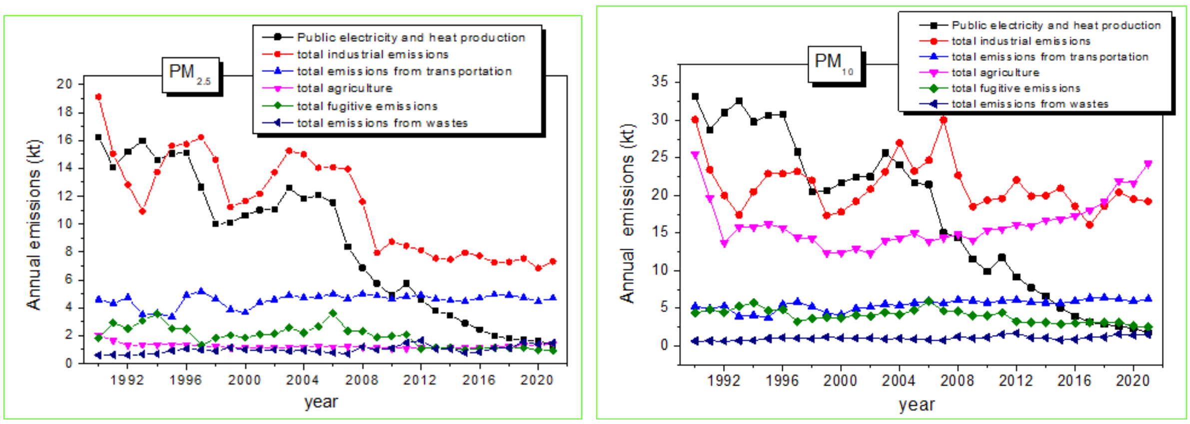

Fig. 3. Annual emissions of PM2.5 and PM10 in Romania, between 1990 – 2021, for 6 sectors of human activities

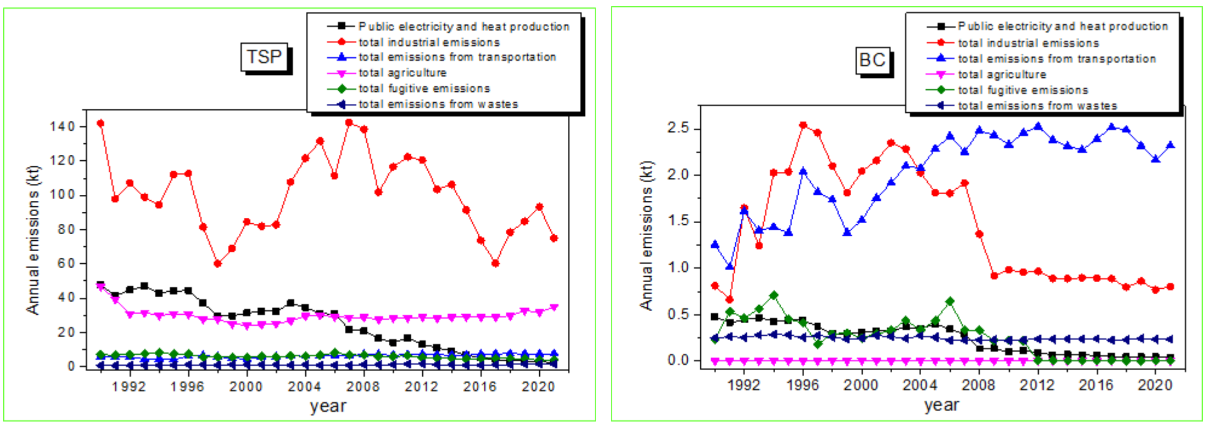

Fig. 4. Annual emissions of TSP and BC in Romania, between 1990 – 2021, for 6 sectors of human activities

There are steady emissions of PM10 from transport and wastes, but they are small. The TSP emissions, there were large fluctuations in the emissions for public energy and heat production, while much lower emissions were recorded from the other sectors, especially from fugitive, wastes and agriculture. As BC is concerned, half of the time corresponds to highest emissions from industry and half from transportation, while the other types of emission sources have small contributions. From 2009, there are steady emissions of BC from transportation.

Fig. 5 presents a comparison between the annual emissions of all four classes of suspended particles considered in this paper, only from the industrial processes, while fig. 6 presents a similar comparison from fugitive emissions and agriculture, and fig. 7 shows the comparison of emissions from waste management.

The annual emissions from public energy and heat production respect the following inequality: TSP > PM10 > PM2.5 > BC, the latter emissions being very low as compared to the others. There are similar trends for TSP, PM10 and PM2.5 and their emissions present a substantial decrease, from 2011. The highest emissions from industry are of TSP, while the lowest are for BC. Also, one can notice that there are similar trends for PM10 and PM2.5 emissions from industry and they are rather steady. The annual emissions from fugitive sources respect the same inequality as from public energy and heat production.

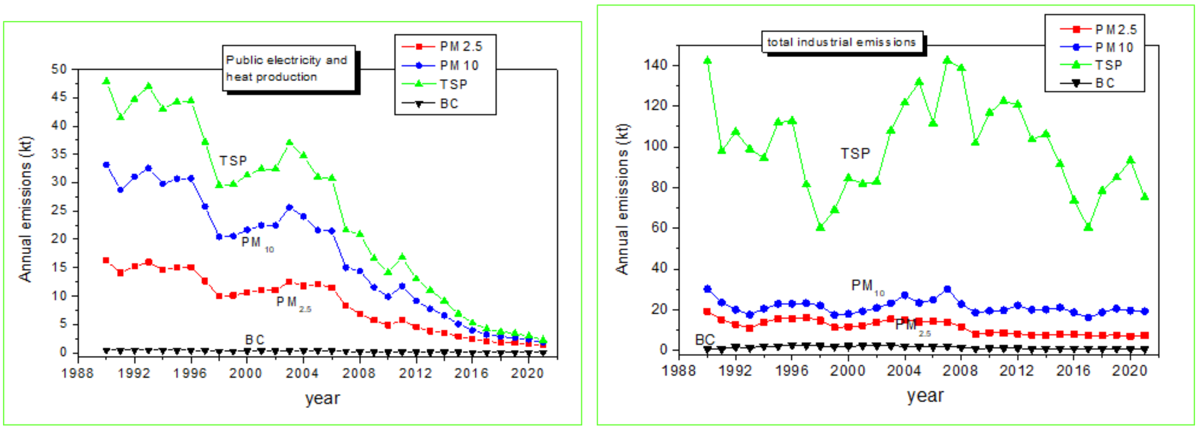

Fig. 5. Comparison between the annual emissions of all 4 classes of suspended particles, for Romania, between 1990 – 2021, from 2 sectors of human activities:
1. Public electricity and heat production; 2. Industry

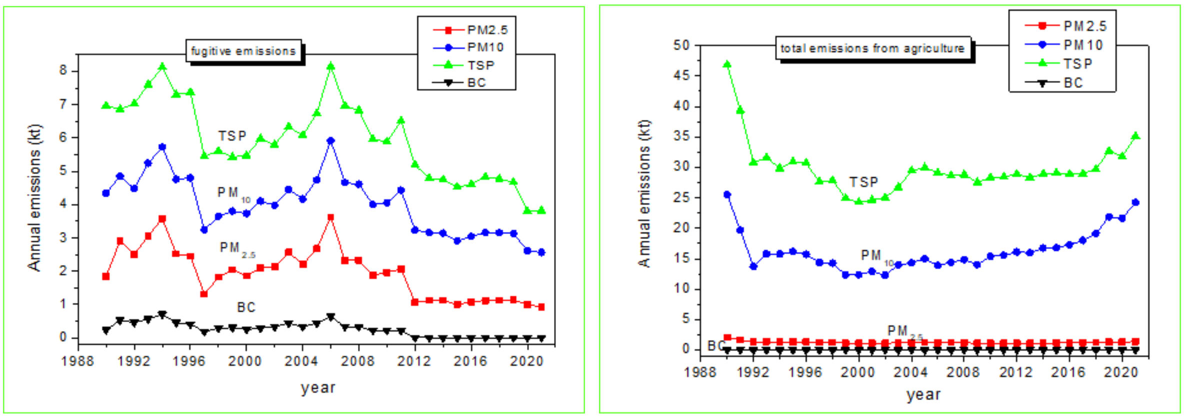

Fig. 6. Comparison between the annual emissions of all 4 type of suspended particles, for Romania, between 1990 – 2021, as fugitive emission and from agriculture

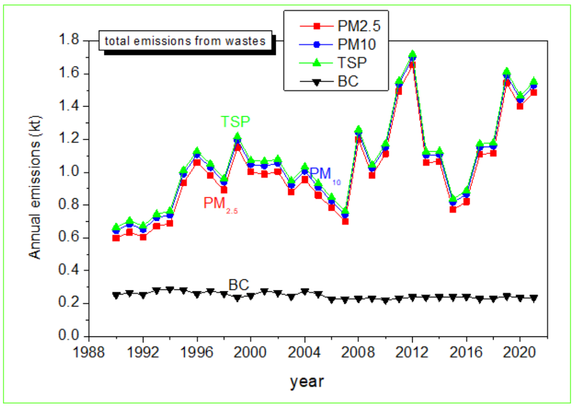

Fig. 7. Comparison between the annual emissions of all 4 type of suspended particles, for Romania, between 1990 – 2021, from wastes

The emissions of TSP, PM10 and PM2.5 from fugitive sources present similar trends, and the emissions are steady from 2012. As the annual emissions from agriculture are concerned, the highest are of TSP, then PM10, while PM2.5 and BC emissions are low. There are similar trends for TSP and PM10 from agricultural activities. Wastes determined similar trends (increasing, in average, from 2014) for TSP, PM10, and PM2.5, while BC emissions are low and steady.

It was considered relevant to assess and compare the annual emissions of PM2.5, PM10, TSP and BC from the transportation subtypes and from the agricultural subtypes of activities, and the results are presented in figs. 8 and 9. It can be seen that, for PM2.5, the highest emissions were given from road transportation and the lowest from aviation, while, in general, there were higher emissions from livestock than for other agricultural activities (i.e. crops). For PM10, the highest emissions from transportation were recorded from railways, followed by road activities and livestock. There is an obvious continuous increase in emissions of PM10 from railways from 2010. In the case of TSP, the highest emissions were recorded, between 1990 and 2021, in Romania, from livestock, followed by farm-level agriculture (crops), then from road transportation. There is an increasing trend of TSP emissions both from farm-level agriculture and road transportation. Finally, for BC, road transportation gave the highest emissions from all types of transportation modes, while the other emissions sources contribute insignificantly.

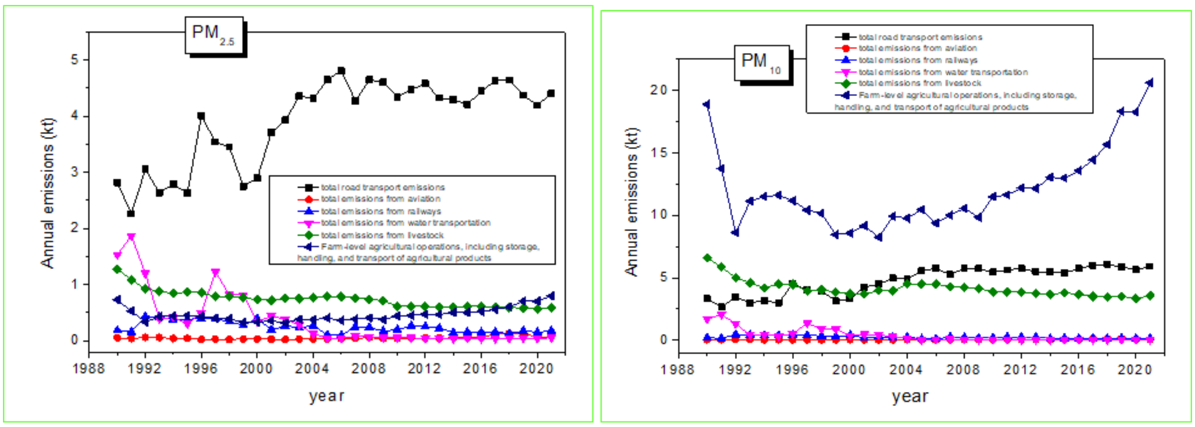

Fig. 8. Annual emissions of PM2.5 and PM10 in Romania, between 1990 – 2021, from transport subtypes and from agricultural subtypes of activities

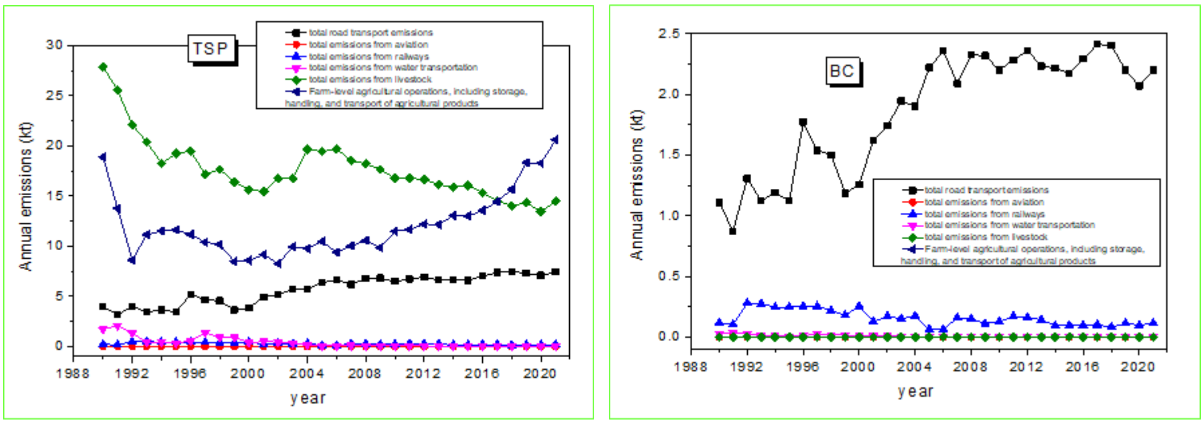

Fig. 9. Annual emissions of TSP and BC in Romania, between 1990 – 2021, from transport subtypes and from agricultural subtypes of activities

Since Romania has still important agricultural activities, it was considered useful to compare the annual emissions of all 4 classes of particles from the 2 main subsectors of Agriculture, namely crop management and livestock, respectively. The results are presented in figs. 10. In what concerns the annual emissions from farm-level agriculture (crops), PM10 and TSP practically overlap and have much higher emission values than the other types of particles. As for the emissions from livestock, TSP had the highest emission, followed by very far by PM10.

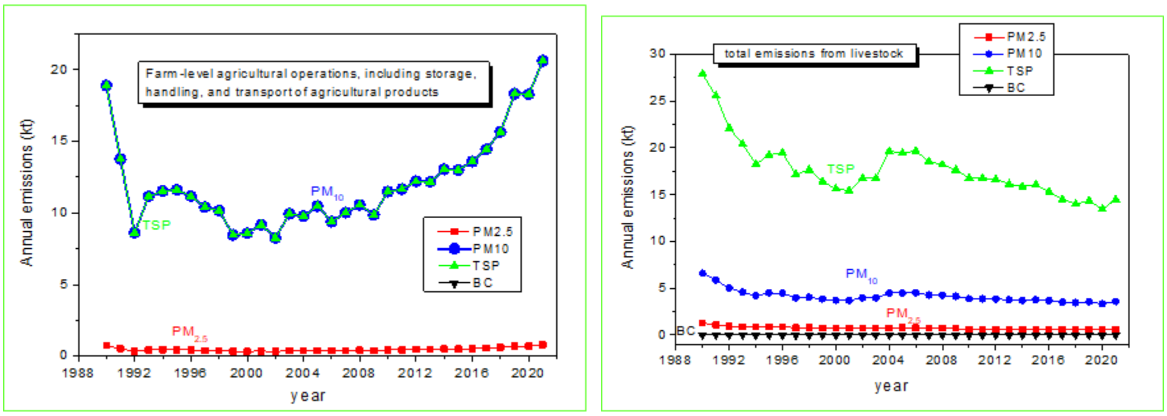

Fig. 10. Annual emissions of all 4 classes of particles, in Romania, between 1990 – 2021, from 2 subtypes of agricultural activities: i) crop management, including their storage, handling and transport; ii) livestock, mainly manure management

Finally, the emissions of particles from the 4 types of transportation modes are presented in Fig. 11. As the annual emissions from aviation are concerned, PM2.5 and PM10 practically overlap and have much higher emission values than the other types of particles (TSP and BC). The annual emissions from railways have very similar values and trends for TSP, PM10 and PM2.5, while they have lower values for BC.

From the analysis of the data in fig. 11, it can be noticed that the emissions from water transportation present very similar values and trends for TSP, PM10 and PM2.5, while much lower values were recorded for BC. All four classes of particles present a steady emission from water transportation from 2008, as it was also the case with the emissions from road transportation. Instead, in the case of road emissions, the emitted quantities of particles and can be ordered as following: TSP > PM10 > PM2.5 > BC, for the latter much lower emissions being recordered.

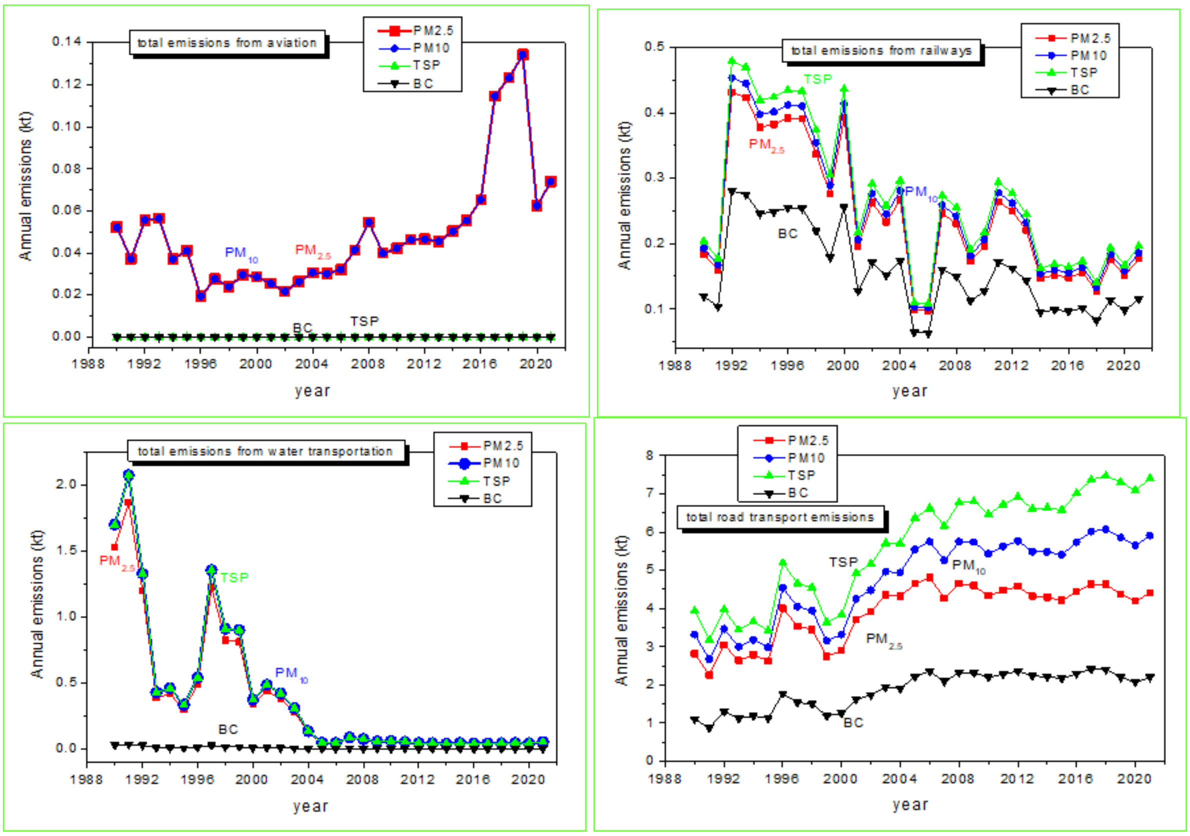

Fig. 11. Annual emissions of all 4 classes of particles, in Romania, between 1990 – 2021, from 4 separate types of transportation means: aviation, railways, on water, on roads

4. CONCLUSIONS

This paper presented an analysis of the annual emissions in Romania, between 1990 and 2021, of four classes of particles suspended in the atmosphere, from different anthropogenic activities. It could be noticed that each sector of activity, either industry or agriculture, transportation, waste management or from fugitive sources contribute in a specific manner to the emissions of PM2.5, PM10, TSP and BC. At their turn, different subtypes of human activities, such as transportation modes or agricultural activities, must be assessed individually with respect to their contribution to pollution, especially in the case of particulate matter, since their evolution are not similar, nor closely correlated, as thus, not predictable in all, but in part.

Given the fluctuating evolution of the economy and finances, of politics, but also of the climate, the evaluation of the pollution trends must be made continuously, and correlated with all the influencing factors and with those they influence, in turn, especially weather, climate and human health.

The impact of particulate matter on human health has led to increased public awareness and advocacy for cleaner air and more sustainable practices to reduce air pollution. Mitigating the impact of particulate matter involves a combination of regulatory measures, technological and awareness initiatives.

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