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AIR EMISSIONS INVENTORY FROM A ROMANIAN CONSTRUCTION MATERIALS FACTORY

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Abstract

The paper aimed to present the main methodologies for estimating atmospheric emissions, with an example for a construction materials factory. It is based on the statistical data provided by a pre-dosed dry mortar factory, for which both the emissions specific to the limestone quarry where the raw material comes from, as well as the emissions from the manufacturing process, were analysed. The presented method, used for the elaboration of the emission inventory and not only, requires the use of specific emission factors for each emission-generating activity in an area or in a company. The method can be used for other purposes as well, including for calculating taxes to the Environmental Fund. Sometimes, due to legislative oversights or the impossibility of including a source in the existing list, it is up to the evaluator to choose the most correct approach.

***Key words**: air pollutants, emission source, emission factor, emission inventory, Environmental Found.*

INTRODUCTION

Substances emitted into the atmosphere by human activities and natural sources represents one of the main causes of many current and potential environmental problems, including acidification, eutrophication, and tropospheric ozone pollution, air quality degradation, global warming, damage of buildings and other structures, human and ecosystem exposure to hazardous substances (EMEP/EEA, 2019).
Air pollution is a major concern and impacts the quality of human life and the smooth functioning of human activities. In addition, the almost rampant industrialisation that has taken place in recent decades has visibly affected the quality of the air we breathe, with a significant proportion of the world's population suffering from pollution by various harmful substances and particulate matter in the air (Ciobanu C. et al., 2021). The rapid growth of urban complexes has increased pollution emissions from residential constructions, transportation, power generation and residential (Nastase G. et al., 2018).
For this reason, it is necessary to have quantitative information on these air emissions and their sources. And then, based on the data and information obtained, to be able to: define environmental priorities and identify the activities responsible for the problems; assess the potential environmental impacts; evaluate the costs to solve the identified problems as well for air pollution strategies and plans; establish the targets are being achieved. Air pollutant emission inventories are an essential basis for controlling and managing pollutant emissions (Xu B. et al., 2020).
Therefore, at the EU level there had to be a unified approach and a methodology for estimating atmospheric emissions from different types of sources, thus creating what is known today as an emissions inventory (Figure 1). Representative emission inventories are crucial for air quality applications as they will largely determine the accuracy of subsequent air quality modelling results (Georgiou et al., 2020).

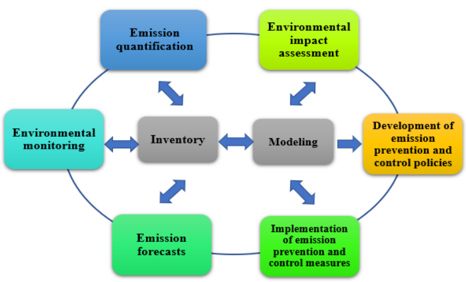

Figure 1. The role of emission inventories for air quality assessment

Emission inventories are now considered indispensable tools for a wide range of environmental measures, such as chemicals management and air pollution prevention.

The work on the original European monitoring and evaluation programme (known as CORINAIR) started in 1985. It started from the idea that it is not possible to measure air emissions from all the individual source's type. Therefore, it was elaborated a basic model based on statistic data about the main industrial activities (location, production rate, operating condition etc.), their typical sources and pollutants and emission measurements over a period.

A guidebook has since been developed and maintained by the UNECE Task Force on Emission Inventories and Projections (TFEIP) under the Convention on Long Range Transboundary Air Pollution (LRTAP Convention).

The guidebook continues to be published and updated by the European Environment Agency (EEA) and it is known as *EMEP/EEA Air Pollutant Emission Inventory Guidebook*, its latest version being the one from 2019 (EMEP = European Monitoring and Evaluation Programme). Periodically, the European Environment Agency publishes the results of the annual reports made by the member countries.

MATERIALS AND METHODS

The structure and content of local emission inventories must meet two essential criteria: to allow the use as input data in mathematical models of pollutant dispersion; to include all the sources of atmospheric pollutants existing in the area for which the inventory is drawn up. In order to develop an emission inventory for an area or company with specific activities one must:

- list the types of sources;
- determine the type of air pollutant emission from each of the listed sources;
- examine the literature to find valid emission factors for each of the pollutants of concern;
- determine the number and size of specific sources;
- calculate the total emissions (Valero D.A., 2008).

The EMEP/EEA Guidebook has two key functions: to provide procedures that enable users to develop emission inventories that meet quality criteria for transparency, consistency, comparability and accuracy; to provide estimation methods and emission factors that are applicable for various degrees of complexity.

Romania is obliged to report annually to the EEA the inventories regarding pollutant emissions into the atmosphere, in accordance with the provisions of European legislation and international conventions in the field to which it is a party. Economic operators are obliged to report the data specific to their activities, so that the environmental protection authorities can centralise them and report them in turn for the annual emission inventory.

The procedure for making local emissions inventories and the national inventory in accordance with the requirements of the EMEP/EEA Guidebook is provided in the Ministry Order no. 3299/2012 for the approval of the methodology for making and reporting the inventories regarding the emissions of pollutants into the atmosphere.

Although the guide covers a wide range of activities, for which useful information is specified, it cannot cover the millions of possible situations. Therefore, often those who are involved in reporting face difficulties regarding the framing of the activity for which they are reporting, identifying the appropriate emission factors, etc. In such situations, those interested must ensure that they make the best choice using the guide or must look for other methods proposed by countries outside the EU and justify the choice made.

An alternative solution for such situations, quite often used, is the *AP-42 Compilation of Air Pollutant Emissions Factors*, that has been published starting with 1972 as a compilation of US EPA's emissions factor information (US EPA AP-42, 2009). It contains emissions factors and process information for more than 200 air pollution source categories and is constantly updated, like the EMEP/EEA Guidebook.

On the other hand, depending on the estimates made for the emissions inventory, economic agents must also pay taxes to the Environment Fund Administration, for a few types of pollutants.

RESULTS AND DISCUSSIONS

The estimation approach is to combine information on the extent to which a human activity takes place (called activity data or AD) with coefficients that quantify the emissions or removals per unit activity, called emission factors (EF).
The basic equation is therefore:

Emissions = AD x EF

The emission factors are given for three tiers and a tier represents a level of methodological complexity. The three tiers provided by the EMEP/EEA Guidebook (EMEP/EEA, 2019) are the follows:
- Tier 1 is the simplest method that represents a default relation between activity data and emission factor; the activity data are based on statistical information for all the sources and substances;
- Tier 2 is more advanced method that uses more specific emission factors, using country specific information on process specific conditions;
- Tier 3 is the method that uses the latest scientific knowledge in more sophisticated approaches and models; it is recommended where facility-level emission data are available and depends on the specific national circumstances.

Tier 1 is the most used and convenient method as long as there is no complete and available data for Tier 2 and Tier 3.
According to the EMEP/EEA Guidebook, the pollutant emissions estimates are divided into sectors (energy, industrial processes and product use, agriculture, waste, natural sources) and sources are divided into categories and subcategories, each one being assigned an NFR code (Nomenclature For Reporting).
The emission inventories prepared according to the Guidebook are different purposes: providing information to policymakers in the Member States of the European Union, as well as the public; defining environmental priorities; air quality assessment for current situation; development, implementation and updating of plans and programs for air quality management; estimating the specific environmental costs of different policies. Also, the national emission inventories allow to model the dispersion of pollutants at a local scale and to assess compliance by Parties with their emission obligations under the existing international protocols.
The case study presented in this paper refers to a dry mortar factory, for which data were available regarding the specifics of the activities, production capacities, etc. The factory's products are represented by mortars, gypsum- and lime-based facades renders, adhesives, plasters and other products used in construction.
Because the main raw material used in the production process is limestone, when inventorying the sources of emissions, we also considered those specific to the extraction activities from the quarry (the quarry belongs to the same company).
The main phases of the technological process in the limestone quarry are the following:
- detonation, for which nitrammonium (94.5% ammonium nitrate + 5.5% diesel) is used as an explosive;
- transportation of raw limestone by vehicle for processing;
- preliminary sorting of the raw limestone, with the aim of separating the sterile material;
- crushing with the help of jaw crushers and final sorting with the help of vibrating sieves;
- temporary storage of the resulting sorts (40 - 80 mm, 80 - 110 mm and over 110 mm).

The local transport of the granular material inside the sorting and crushing station, respectively for temporary storage is done with the help of conveyor belts.
Next, limestone with grains up to 80 mm is transported to the dry mortar factory where, before being processed, it is dried; for this purpose, a dryer is used with a drying capacity of about 50 t/h, with a maximum gas consumption of 250 mc/h. After drying, the limestone is subjected to a process of processing and fine sorting, so that various sorts with granulometry corresponding to the various types of manufactured products are obtained.
The local transport of dry limestone, the making of mixtures and the packaging of finished products are operations that take place in closed spaces, where the existed exhaust pipes are provided with batteries of filters with bags, to

limit the emission of solid particles into the atmosphere.

The emission from detonation depends on the chemical composition of explosive and on the weight of explosive used. Therefore, the main pollutant generated from explosives detonation is the carbon monoxide. In second place in terms of the quantity generated are the particulates; such large quantities of particulate are generated in the shattering of the rock and earth by the explosive that the quantity of particulates from the explosive charge are not significant and cannot be distinguished. Nitrogen and sulphur oxides are also generated, but only limited data are available on these emissions.

Emissions of particulate matter (PM), PM-10, and PM-2.5 occur from specific operations in limestone quarrying and processing. A substantial portion of these emissions consists of heavy particles that may settle out within the plant. The main variables that affect uncontrolled PM emissions are wind and material moisture content. If these are added other variables such as: crusher type, size reduction ratio, fines content etc. Therefore, the emission factors developed from the emissions data gathered at limestone processing facilities are considered representative and were established based on the processed limestone quantity. However, only the emission of particulate matter is considered to be the main type of emission.

Regarding the limestone drying in the mortars factory, are taken into account all the specific emissions from combustion process of the natural gas: black carbon (BC), methane (CH_4), carbon monoxide (CO), carbon dioxide (CO_2), nitrous oxide (N_2O), ammonia (NH_3), nitrogen and sulphur oxides, non-methane volatile organic compounds (NMVOC), heavy metals and persistent organic pollutants. The emissions are calculated based on gas consumption. In Table 1 are presented the type of sources and emissions as well as the bibliographic sources from which the emission factors were taken for the analysed case study.

Table 1. Sources of pollution and types of specific emissions

Sources categories	NFR code	Type of emissions	Emission factors
Detonation of explosive (ammonium nitrate with 5 – 8% fuel oil)		products of explosives use (CO, NO_x, SO_2)	US EPA: AP 42, Fifth Edition Compilation of Air Pollutant Emissions Factors, Volume 1: Stationary Point and Area Sources, Chap. 13.3 - Explosives Detonation, Table 13.3.1
Row limestone loading-unloading		particulate matter	US EPA: AP 42, Fifth Edition Compilation of Air Pollutant Emissions Factors, Volume 1: Stationary Point and Area Sources, Chap. 11.19.2 - Crushed Stone Processing and Pulverized Mineral Processing, Table 11.19.2-1
Processing operations (sorting, crushing, belt transportation)		particulate matter	
Non road mobile machinery (excavators, loaders, off-highway trucks, graders)	1.A.2.g.vii	exhaust emissions	EMEP/EEA Air Pollutant Emission Inventory Guidebook, 2019, Chap. 1.A.4 - Non road mobile machinery, Table 3-1
Limestone drying (gaseous fuels)	1.A.2	emission from combustion process (CO, NO_x, SO_x, particulate matter, etc.)	EMEP/EEA Air Pollutant Emission Inventory Guidebook, 2019, Chap. 1.A.2 - Manufacturing industries and construction (combustion), Table 3-3

As stated in Table 1, the methodologies proposed by the European guide and the American standard were considered.

It is important to mention that special attention must be paid to the measurement units when choosing the emission factor values, especially in situations where the sources use different measurement unit systems.

In Table 2, in which the emission factors are presented, units from the International System of Units of Measure were used.

Empty cells mean that either that type of pollutants are not generated from the respective activity, or that no emission factors are established for some pollutants (these pollutants are considered insignificant from a quantitative point of view).

Table 2. Emission factors taken into account

Source	limestone quarry							factory	
	Detonation of explosive	Row limestone loading	Row limestone unloading	Screening	Crushing	Conveyor transport	Non road mobile machinery	Limestone drying	Non road mobile machinery
Pollutant	[kg/t explosive]	[kg/t limestone]					[kg/t diesel]	[g/Gj]	[kg/t diesel]
BC							1.306	0.03	1.306
CH_4							0.083		0.083
CO	34						10.774	29	10.774
CO_2							3,160		3,160
N_2O							0.135		0.135
NH_3							0.008		0.008
NO_x	8						32.629	74	32.629
SO_2	1							0.67	
NMVOC							3.377	23	3.377
TPM		$5x10^{-5}$	$8x10^{-6}$	0.15	0.0195	0.0015	2.104	0.78	2.104
Cadmium							$0.01x10^{-3}$	$0.00091x10^{-3}$	$0.01x10^{-3}$
Copper							$1.7x10^{-3}$	$0.0026x10^{-3}$	$1.7x10^{-3}$
Chromium							$0.05x10^{-3}$	$0.013x10^{-3}$	$0.05x10^{-3}$
Nickel							$0.07x10^{-3}$	$0.013x10^{-3}$	$0.07x10^{-3}$
Mercury								$0.54x10^{-3}$	
Arsenic								$0.1x10^{-3}$	
Selenium							$0.01x10^{-3}$	$0.058x10^{-3}$	$0.01x10^{-3}$
Zinc							$1x10^{-3}$	$0.73x10^{-3}$	$1x10^{-3}$
Lead								$0.011x10^{-3}$	
POPs							$3.32x10^{-3}$	$5.8x10^{-6}$	$3.32x10^{-3}$

The air emission calculation to be presented below was developed for an average month, in which the activity data (AD) considered are the following: detonation and processing of 56,000 t limestone; using 27 t diesel for non road mobile machinery; using 32,760 mc gas for drying (assuming that around 45% of the extracted limestone is processed in the factory, the rest being sold in various forms to third parties); using 4 t diesel for non road mobile machinery. The quantities of emissions, expressed in kg, are presented in Table 3. Five decimals were kept, so that the highest values could be highlighted in comparison with the insignificant ones, almost zero.

Table 3. Emission of pollutants for an average month

Source / Pollutant	Detonation of explosive	Row limestone loading	Row limestone unloading	Screening	Crushing	Conveyor transport	Total non road mobile machinery	Limestone drying	Total emission [kg]
BC							39.5979	0.03909	39.63701
CH_4							2.5166	0.00000	2.51656
CO	262.8						326.6677	37.78309	627.20277
CO_2							95,811.2000	0.00000	95,811.20000
N_2O							4.0932	0.00000	4.09320
NH_3							0.2426	0.00000	0.24256
NO_x	61.8						989.3113	96.41202	1,147.54730
SO_2	7.7							0.87292	8.60092
NMVOC							102.3906	29.96590	132.35654
TSP		3	0.448	5880	764.4	58.8	63.7933	1.01623	6,771.25751
Cadmium							0.0003	0.00000	0.00030
Copper							0.0515	0.00000	0.05155
Chromium							0.0015	0.00002	0.00153
Nickel							0.0021	0.00002	0.00214
Mercury								0.00070	0.00070
Arsenic								0.00013	0.00013
Selenium							0.0003	0.00008	0.00038
Zinc							0.0303	0.00095	0.03127
Lead								0.00001	0.00001
POPs							0.1007	0.00001	0.10067

Analysing the figures resulting from the calculations, it can be seen (Figure 2) that the largest quantity of emissions is represented by carbon dioxide (91.65%) and total suspended particles (6.48%).

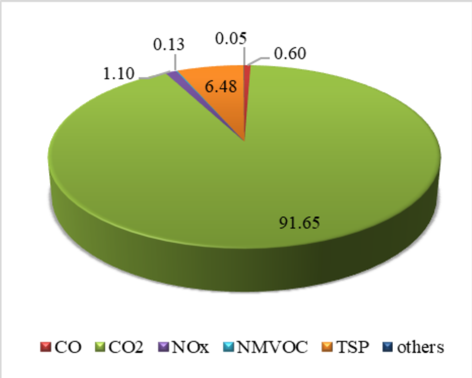

Figure 2. Main air emissions (in percentage)

The main source of CO_2 is the combustion process specific to the non road machinery, that depends on the diesel consumption. Carbon dioxide is not considered a pollutant, as it occurs naturally in the air. However, it is a greenhouse gas and the increase in greenhouse gas emissions causes warming the climate of our planet which is an actual and important environmental problem.

On the other hand, the particulates are mainly resulting from the row limestone processing steps and from the combustion processes which are taken in the engine of the non road machinery.

Also, it can be easily observed that the specific emissions from the quarry activities are significantly higher than those from the limestone drying process. It must be realised that an uncontrolled source, like crushing and sorting processes, will emit at least 10 times the number of pollutants released from one operating properly with air pollution control equipment installed (Valero Daniela A., 2008). It is true that not all the extracted limestone is processed in the factory, but even so, the sources from the quarry have a clearly higher weight.

Last but not least, it should be remembered that, according to the Romanian legislation (MO no. 578/2006), the economic operators owning stationary sources, the use of which affects environmental factors, are obliged to pay taxes to the Environmental Fund for the generated atmospheric pollutant emissions.

Stationary sources that generate pollutant emissions into the atmosphere mean constructions, machines, installations, including ventilation, other fixed works that generate or through which polluting substances are discharged into the atmosphere, as well as fixed combustion installations belonging to the same economic operator, with a power installed total greater than or equal to 1 MW. It can be observed that for the considered stationary sources the monthly amount of taxes isn't so significant, and it is mainly caused by particle emissions (Table 4).

Table 4. The amount of taxes for estimated air emissions

	Total emissions from stationary sources [kg]	Taxes [lei/kg]	Total taxes [lei]
NO_x	96.412020	0.04	3.86
SO_2	0.872920	0.04	0.03
TSP	6645.416230	0.02	132.91
Cadmium	0.000001	16	0.00
Mercury	0.000704	20	0.01
Lead	0.000014	12	0.00
POPs	0.000008	20	0.00
Total			136.81

As can be seen, the relatively small taxes that the company has to pay to the Environmental Fund, due to the atmospheric emissions generated, are not so high as to stimulate the reduction of these emissions.

CONCLUSIONS

National inventories are drawn up in order to fulfil the reporting obligations deriving from the status of a member state of the European Union, respectively, a Party to international conventions, serving, mainly, to draw up national and international policies for the protection of the environment.

The purpose of this article was not to assess the impact on the environment, nor the assessment of the impact on air quality, because of the activities specific to the production of dry mortars.

The intention of the authors was to draw attention to some difficulties encountered by environmental consultants and companies that have to choose the most suitable methods for estimating atmospheric emissions from various sources.

The use of emission factors to estimate the quantities of atmospheric pollutants can be applied in several cases: assessment of the impact on the environment in the design phase; evaluation of the effects on the environment during the operation phase, especially when measurements cannot be made at the source; making annual reports for the emissions inventory; calculation of taxes at the Environmental Fund for atmospheric emissions. However, this method used for the emission inventory does have some errors. The other alternatives are expensive and subject to their own errors: the first alternative refers to monitoring every major source; the second method would be to monitor the imissions at many points at the limit of the property.

The use of emission factors to quantify the quantities of pollutants emitted into the atmosphere from different anthropogenic activities can be applied in different situations: environmental impact assessment, in the design phase; evaluation of the effects on the environment during the operation phase, for situations where measurements cannot be made, and when the estimated values can be used both for the emission inventory and for the fees to the Environmental Fund.

For that reason, there should be and maintain a greater concordance between the provisions of the slightly older legislation and the new requirements arising from the repeated changes of the European legislation, including the Romanian legislation.

For the national emissions inventory, the following must be ensured:

- data transparency, which requires the provision of sufficient documentation so that the method of estimating emissions for each individual source can be followed;
- consistency of the data, which assumes that the methods are in accordance with the best practices and that they are used consistently for all pollutants;
- data comparability, which implies the use of the best methodologies for estimating and reporting emissions;
- completeness of the data, respectively the inclusion in the inventory of all categories of sources/activities, from the entire national territory;
- data accuracy, which implies the use of best practices for estimating emissions according to the provisions of the latest version of the EMEP/EEA Guide.

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