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On the source inversion of fugitive surface layer releases. Part II. Complex sources

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HIGHLIGHTS

- A methodology is proposed to estimate fugitive PM emissions from complex sources.
- Overlapping and/or mobile emission sources are considered.
- Studied sources include aggregate handling and transport on unpaved roads.
- A series of field experiments were conducted under real operating conditions.
- Operation-specific PM_{10} emission factors for complex fugitive sources are provided.

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ABSTRACT

The experimental measurement of fugitive emissions of particulate matter entails inherent complexity because they are usually discontinuous, of short duration, may be mobile, and are affected by weather conditions. Owing to this complexity, instead of experimental measurements, emission factors are used to inventory such emissions. Unfortunately, emission factor datasets are still very limited at present and are insufficient to identify problematic operations and appropriately select control measures. To extend these datasets, a source inversion methodology (described in Part I of this work) was applied to field campaigns in which operation-specific fugitive particulate matter emission factors were determined for several complex fugitive sources, some of which were mobile. Mobile sources were treated as a superposition of instantaneous sources. The experimental campaigns were conducted at ports (bulk solids terminals), aggregate quarries, and cement factories, encompassing powder handling operations and vehicle circulation on paved and unpaved roads. Emission factors were derived for the operations and materials involved in these scenarios and compared with those available in the emission factor compilations. Significant differences were observed between the emission factors obtained in the studied handling operations. These differences call into question the use of generic emission factors and highlight the need for more detailed studies in this field.

1. Introduction

Fugitive emissions, as defined by the US regulations (title 40 of the Code of Federal Regulations, sections 70.2 and 71.2), denote a broad category of "emissions which could not reasonably pass through a stack, chimney, vent, or other functionally-equivalent opening". This definition by exclusion reflects the variety and complexity of fugitive sources. Of the pollutant fugitive sources, particulate matter (PM) sources possibly exhibit the greatest complexity.

Indeed, though standard methods for the direct experimental quantification of channelled PM emissions are available (e.g. ISO 9096), which allow accurate and relatively simple routine control, this is not the case with fugitive emissions, probably because of the inherent complexity entailed in fugitive PM quantification and control, owing to different factors:

- Fugitive PM is transported from its origin by fluctuating wind, rather than at a constant flow rate (as is the case in channelled emissions).
- Almost all fugitive PM emission-generating industrial activities are of a discontinuous nature and short duration, and emission frequency and intensity can vary even within a workday.

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- Source position can vary with time – sometimes the source moves continuously along an essentially arbitrary path –.
- Fugitive PM emission rates are often affected by weather conditions (wind speed and direction, atmospheric stability, etc.).
- These operations are often carried out by workers, which introduces a human factor.

Furthermore, dust from one source may become mixed with that from others, because each activity usually involves several overlapping operations, which do not necessarily occur in a particular, well-defined sequence. For example, in many bulk solids processing industries, bulk solids are often stored in the open air. This activity usually encompasses several operations: arrival at the bulk solids reception area, piling to form heaps or mounds, transport by a shovel truck or similar vehicle, and finally discharge or unloading of the material for dispatch or subsequent processing. Each of these operations can produce fugitive PM emissions of varying magnitude (Monfort et al., 2011).

Consequently, while channelled PM emissions can be inventoried by experimental measurements at source, fugitive emissions are estimated by means of emission factors (EFs). EFs estimate the PM emission rate based on a unit magnitude that quantifies the intensity of the operation: that is, the emissions are assumed to be directly proportional to that magnitude. In practice, EFs for bulk solids handling are considered to be proportional to the mass of processed material, whereas EFs for vehicle traffic are expressed per unit distance travelled.

At present, there are a number of fugitive PM EFs. The most widely used are those set out in the US Environmental Protection Agency (US EPA) AP–42 compilation (US EPA, 1995; Section 13.2). These fugitive PM EFs are classified into several categories, viz.: (i) paved roads, (ii) unpaved roads, (iii) aggregate handling and storage piles, and (iv) industrial wind erosion. All take the form of predictive empirical equations that depend on a few explanatory experimental parameters.

The AP–42 paved roads PM emissions formula was originally developed by Cowherd et al. (1974), this being revised to incorporate additional tests (US EPA, 1995; Section 13.2). Paved road dust emissions are thought to be one of the main contributors to urban PM pollution (Pant and Harrison, 2013; Amato et al., 2013). Possibly because of this, the determination of paved road PM EFs has been a subject of extensive research (Claiborn et al., 1995; Venkatram et al., 1999; Abu-Allaban et al., 2003; Etyemezian et al., 2003; Ketzel et al., 2007; Amato et al., 2010; among many others).

In contrast, fugitive PM emissions belonging to the other AP–42 categories have drawn much less attention. For example, the category aggregate handling and storage piles is used to represent a very extensive array of operations and materials. Despite such a wide scope, the number of test data considered to derive the EF predictive equation is somewhat limited. In particular, the current formula to estimate these emissions stems from Muleski et al. (1987), which encompasses the results obtained in three test reports that involved coal dumping in a coal-fired power plant, drop of prilled sulfur, and loading of fly ash into open trucks, respectively.

Since the original work by Cowherd and co-workers (Cowherd et al., 1974, 1979; Muleski et al., 1987), relatively few additional studies aimed at deriving EFs for aggregate handling fugitive PM sources have been conducted (Vrins et al., 1994; Muleski et al., 2005; Martín et al., 2007; Hosseini and Stockie, 2016). The scarcity of information is also observed in the EFs set out in the European Environment Agency (EEA) air pollutant emission inventory guidebook (EEA, 2016, Chapter 2.A.5.c) used in European inventories, which contains only generic EFs for these emissions (Table 1).

However, greater detail (in terms of a specific EF for each operation involved in an activity) is deemed of great interest, not just in order to be able to estimate the emissions more accurately but also to be able to identify the most problematic operations and to establish appropriate corrective measures. For example, in a facility's design phase, it would be interesting to be able to determine which facility layout gave rise to the least emissions. This can hardly be done with current information.

Part I of this work (Sanfélix et al., 2015) describes a mathematical framework for the estimation of fugitive emissions. The framework consists of a dispersion model that is flexible enough to deal with the complexity of fugitive PM sources. Part II is a follow-on study in which the model is applied to field campaigns determining specific fugitive EFs for several complex fugitive sources: raw materials loading and unloading at bulk solids wharves, truck circulation on unpaved roads, and raw materials handling with shovel trucks. These sources were studied under actual operating conditions. Sometimes several sources, some of which were mobile, were concurrently involved.

2. Calculations

2.1. Source inversion calculations

The calculations performed to obtain the EFs required solving an inverse problem (Isakov, 1990), described in detail in Part I of this work (Sanfélix et al., 2015). The problem basically consisted of calculating the emission rate of an array of pollutant fugitive sources, having determined the concentrations of these pollutants at a (usually limited) number of points in the source surroundings.

The methodology proposed in Part I consisted of solving the problem in two steps. In the first, the pollutant concentration fields were calculated, assuming a unit emission, by means of an atmospheric dispersion model. The proposed model consisted of the numerical solution of the transport equation, which was an equation in partial derivatives solved by the finite volume method. The second step involved the solution of a linear regression problem. Using the superposition principle (Carslaw and Jaeger, 1959), the concentrations at a given point were expressed as a linear combination of those calculated separately for each source. The unknown linear coefficients were the emission rate estimates, obtained by linear least squares fitting.

To quantify the goodness of fit and verify the robustness of the obtained EFs, a bootstrap technique was used (Efron and Tibshirani, 1993). Since the EFs were derived from autocorrelated data (concentration time series), bootstrap replicates were constructed by randomly selecting non-overlapping (12-min long) blocks with replacement among the observations (Künsch, 1989). Furthermore, the least squares method used involved a subjective component through the definition of a threshold concentration, below which the concentrations were not considered in the sum of squared residuals (see Part I, Sanfélix et al., 2015). To also account for its

Table 1
PM EFs for handling of mineral and metal products (EEA, 2016).

Industry	PM EF ($g\ t^{-1}$)		
	TSP	PM_{10}	$PM_{2.5}$
Mineral products	12	6	0.6
Metal products	4	2	0.2

[TSP] total suspended particles.
[PM_{10}] PM less than 10 μm in aerodynamic size.
[$PM_{2.5}$] PM less than 2.5 μm in aerodynamic size.