

Atmospheric depositions of persistent pollutants: methodological aspects and values from case studies

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Abstract

Deposition monitoring, already in use by government control organizations of various countries, contributes to an important increase in experimental knowledge on pollutant deposition fluxes, on their environmental fate and on the possible effects on human health.

At the European level, the necessity to consider with extreme attention the environmental contamination due to deposition, has brought to adopt a series of legislative measures and recommendations; this has contributed to set up environmental surveillance systems and monitoring campaigns for a series of pollutants which may accumulate in the environment as persistent organic pollutants (POPs) and for metals.

More recently, with DL.vo 155/2010, the necessity to consider, in the development of monitoring stations, the possibility to detect also data on deposition rates which represent a non-direct exposure of the population through the food chain. For sampling the Decree considers only two types of depositions: for total deposition (bulk and Bergerhoff) and wet only deposition.

Key words

- air quality
- monitoring
- legislation deposition
- metals
- persistent organic pollutants

INTRODUCTION

National legislation and European directives define the term total or bulk deposition as “the total mass of pollutants which is transferred from the atmosphere to surfaces (e.g. soil, vegetation, water, buildings, etc.) in a given area within a given time”. This parameter is measured using deposition systems which in general are simply made up of a funnel and a total collecting system. Nevertheless in time more complex apparatuses have been developed which allow to differentiate dry and damp deposition sampling, specific sampling for mercury, directional type of sampling. The determination of the deposition rate generally involves a sampling period for the duration of a week or a month and are carried out to cover the whole year. The deposition is expressed as surface mass for a given time reference, generally μg of pollutant per m^2 per day ($\mu\text{g m}^{-2} \text{d}^{-1}$).

Even considering the possible interferences that such a system may have, for example the introduction in the collecting system of objects not correlated to the deposition of pollutants such as insects, leaves, etc., it has however proven in time to be extremely useful, especially for a series of pollutants which may accumulate in the environment as persistent organic pollutants (POPs), in particular polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF), polychlorinated biphenyls (PCB), and metals. The measurement of pollutant fall back from atmosphere to soil, in environmental surveys and in monitoring networks, is greatly used as it produces important information on the area's contamination state and helps in evaluating the exposure of the general population.

These types of surveys are presently carried out in some areas affected by industrial technological cycle emissions of organic and inorganic micropollutants, as for example: iron and steel plants, thermoelectric coal plants, cement plants, incinerators and storage waste fires. As is well-known, the presence of micropollutants in the emissions from large industrial plants entails a fall out to soil of numerous pollutants: metals and metalloids (As, Cd, Ni, Pb, Tl, V, etc.), polycyclic aromatic hydrocarbons (PAH), PCDD/F, polychlorinated biphenyls dioxin-like compounds (DL-PCB). This causes an appreciable contribution of these substances to soil and to some agricultural produce, in particular to forages, and consequently in the food chain, mainly through milk and dairy products, eggs, meat, fish. Therefore, there is an exposure, of the general population through food assumption which appears to be the principal source of PCDD/F assumption according to the World Health Organization Regional Office in Europe (WHO) and which is calculated in 95% of the total assumption [1].

An assessment of contamination by micropollutants in one area can also be performed using bioaccumulators, such as mosses and lichens, however the use of deposimeters is widely experienced in various countries and some of these have also developed guidelines and limits can be related to possible human exposure.

Several international programmes, among which EMEP (Co-operative Programme for Monitoring and Evaluation of Long-Range Transmission of Air Pollutants in Europe), OSPAR (Oslo and Paris Commission Convention for the Protection of the Marine Environment of the North East Atlantic) and LRTAP (Convention on the Long Range Transboundary Air Pollution Protocol on POPs), foresee the monitoring of metals, PAH, PCDD/F, PCB and DL-PCB, in atmospheric depositions with the aim to deepen the knowledge of the levels of such pollutants in depositions.

REGULATORY AND METHODOLOGICAL ASPECTS

Currently, in several countries specific deposition monitoring programmes are carried out for environmental and sanitation evaluations; this had brought to define and adopt limits or guide values in particular for PCDD/F, DL-PCB and some metals.

At the European level, the necessity to consider with extreme attention the environmental contamination due to deposition, has brought to adopt a series of legislative measures and recommendations. Following the Stockholm Convention on POPs [2] whose scope is to reduce to the minimum their emission, the European Commission, in a Communication has underlined that: "dioxins present in air can deposit on vegetables or in water and from there pass in food and in fish and through food consumption enter the food chain" [3-5]. Directive 2004/107CE has also paid serious attention to population exposure through the food chain underlining that: "the effects of arsenic, cadmium, mercury,

nickel and polycyclic aromatic hydrocarbons on human health, including via the food chain, and the environment as a whole, occur through concentrations in ambient air and via deposition; the accumulation of these substances in soils". Furthermore, it has recommended that member States should promote research on human health and environmental effects of such pollutants in particular due to deposition considering soil deposition flows determination a key factor for food chain contamination control and for human exposure besides being a good environmental control system [6]. In Table 1 levels of metals and PCDD/F in depositions from several European countries are summarized [7-11].

Some European countries have introduced in their legislation rules pertaining to atmospheric pollutant deposition; this has contributed to set up environmental surveillance systems and monitoring campaigns for metals, PCDD/F, DL-PCB, PAH. Belgium, in particular, has acquired a regional guideline for PCDD/F+DL-PCB in atmospheric deposition of 10 pg I-TE m⁻² d⁻¹, and has developed a proposal for a national guideline of 8 pg WHO-TE m⁻² d⁻¹. Such values have been backed-up by some studies which have correlated the PCDD/F concentrations, observed in the atmospheric depositions, with the population's global assumption evaluations. One of the studies has considered various areas in Belgium which have civil and industrial PCDD/Fs output sources and has proposed a guideline which correlates the Tolerable Daily Intake (TDI) with PCDD/F deposimetric data. TDI is the daily amount of pollutant that has been assessed safe for human being on long-term basis, is expressed as pg WHO-TE per kg of body weight (bw). The proposed guideline is reported in Table 2 [12-14].

France has also dealt with the same problem; in the report *Méthode de surveillance des retombées des dioxines et furanes autour d'une unité d'incinération des ordures ménagères (UIOM)* by the Istitut National de

Table 1

Metals in bulk depositions measured in different European sites: rural, urban and industrial [7] and PCDD/F in bulk depositions measured in some European countries [8-11]

Metals in bulk depositions			
Pollutants	Rural areas µg m ⁻² d ⁻¹	Urban areas µg m ⁻² d ⁻¹	Industrial areas µg m ⁻² d ⁻¹
Arsenic	0.082-0.43	0.22- 3.4	2.0-4.3
Cadmium	0.011-0.14	0.16-0.90	0.12-4.6
Nickel	0.03-4.3	5-11	2.3-22

PCDD/Fs in bulk depositions		
Country	Urban sites pg I-TE m ⁻² d ⁻¹	Rural sites pg I-TE m ⁻² d ⁻¹
Belgium	0.9-12	0.7-3.1
Germany	< 0.5-464	7-17
France	0.5-17	1.0-10
Denmark	300 -31 600	300-1700
United Kingdom	0.4-312	Nv-517

PCDD/F: polychlorinated dibenzofurans.

Table 2

Correlation between PCDD/F and DL-PCB in atmospheric deposition and tolerable daily intake (TDI) [12-14]

	Deposition pg WHO-TE m ⁻² d ⁻¹	TDI pg WHO-TE/kg _{bw}
Van Lieshout, <i>et al.</i> [12]		
PCDD/F (annual average)	3.4-14	1-4
PCDD/F (monthly average)	6.8-27	
Cornelis, <i>et al.</i> [13]		
Deposition PCDD/F+DL-PCB (annual average)	8.2	2
Deposition PCDD/F+DL-PCB (monthly average)	21	
LAI (Länderausschusses für Immissionsschutz) [14]		
Deposition PCDD/F+DL-PCB (annual average)	4	1.9-2.3

PCDD/F: polychlorinated dibenzofurans; DL-PCB: polychlorinated biphenyls dioxin-like compounds.

l'Environnement Industriel et des Risques (INERIS) [15] the role played by the atmospheric depositions measurements for environmental surveillance and for the evaluation of micropollutants transfer to the food chain has been underlined. In this report a value guide of 40 pg I-TE m⁻² d⁻¹, starting from a 4 pg I-TE g⁻¹ of dairy products fat contamination, has been suggested; this value is far superior to what foresees by legislation. Other indications regarding atmospheric depositions of PCDD/F [10], are contained in air quality surveillance programmes, in areas where there are incinerator plants, in which it is reported that values lower than 5 pg I-TE m⁻² d⁻¹ may be considered as urban-industrial plot (land, acreage) and values over 16 pg I-TE m⁻² d⁻¹ should be considered as a sensitive human contribution [16]. The latter levels should give way to in-depth sampling analyses and micropollutants congeners profiles studies, in order to identify/evaluate the main sources and activate necessary solutions or mitigate contamination.

Germany has included in its legislation, *Technische Anleitung zur Reinhaltung der Luft-TA Luft* [17], limits for dust deposition both for long periods (350 mg m⁻² d⁻¹ annual media), and limits for selected metals referring to specific norms [18]. Furthermore, it developed

a guideline for PCDD/F+DL-PCB atmospheric depositions, showing a value of 4 pg WHO-TE m⁻² d⁻¹; this value is related to the TDI as already reported for Belgium [12, 13].

Other countries have included in their legislature deposition limits only for dusts and/or metals; *Table 3* summarises the different deposition flux limits adopted in the countries cited above.

Concerning Italy, already in the early 1980s, with Law no. 615 "Measures against atmospheric pollution" of July 13 1966, the "Central Commission against atmospheric pollution" was established by and at the Ministry of Health. Within the Commission worked an *ad hoc* group with the task to develop limits and analytical methods for some air pollutants. In that range there was a proposal (1983) also for values of sediment dusts detected with bulk deposition and expressed in mg/m², divided in 5 ascending classes indicating various acute pollution levels:

- | | |
|-------------------------------|--|
| 1) Virtually absent dustiness | < 100 mg m ⁻² d ⁻¹ |
| 2) Low dustiness | 100-250 mg m ⁻² d ⁻¹ |
| 3) Medium dustiness | 251-500 mg m ⁻² d ⁻¹ |
| 4) Medium-high dustiness | 501-600 mg m ⁻² d ⁻¹ |
| 5) High dustiness | > 600 g m ⁻² d ⁻¹ . |

Table 3Current limit values (annual average) in some European countries for dust deposition (PM = mg m⁻² d⁻¹), PCDD/F + DL-PCB (pg WHO-TE m⁻² d⁻¹) and metals (µg m⁻² d⁻¹) in atmospheric depositions

Country (reference)	PM	PCDD/F+ DL-PCB	As	Cd	Hg	Ni	Pb	Tl	Zn
Austria [32]	210	–	–	2	–	–	100	–	–
Belgium [13, 33]	350 650*	8.2 21*	–	2	–	–	250	–	–
Croatia [34]	350	–	4	2	1	15	100	2	–
Germany [14, 17, 18]	350	4	4	2	1	15	100	2	–
United Kindom [35]	200	–	–	–	–	–	–	–	–
Switzerland [36]	200	–	–	2	–	–	100	2	400
Slovenia [37]	200	–	–	2	–	–	100	–	400

PCDD/F: polychlorinated dibenzofurans; DL-PCB: polychlorinated biphenyls dioxin-like compounds.

* Monthly average.

Such a proposal never went beyond a study level and there was no follow-up in the legislation of the time. Nevertheless the detection method was well applied also in period when reference limits were lacking; it should be kept in mind that one of the first active networks in Italy goes back to 1977. In that year the Istituto Superiore di Sanità (ISS) set up in the Seveso (Milan) area determinations aimed at measuring dioxin fall-out during cleanup operations of an area accidentally contaminated by an incident which occurred at the ICMESA plant. The incident involved a chemical substances leakage, among which tetraclorodibenzo-dioxin (2,3,7,8-TCDD), from a chemical reactor for the production of herbicides. The determination network which was set up, which comprised 16 bulk type deposition apparatuses made up basically by a funnel/bottle collector, contributed effectively to fall-out monitoring of the vast area potentially interested to pollution [19].

Afterwards, in the Decree of May 20 1991 (Criteri for data collection inherent air quality), in Annex 1, item 1.6, Non-automatic measures – Species to analyse, was reported: Atmospheric depositions, which may be dry or damp. Damp depositions normally interest distant areas. In industrial areas dry depositions are significant, but they must not be confused with deposition dust. Deposition dust: it comprises particulate material having a very high granulometry and which sediments due to the action of a gravity field. It is evaluated by means of collecting it in appropriate deposition instruments. On the deposited dust, various kinds of chemical analyses may be conducted.

More recently, with Directive 2004/107/EU, and with the subsequent transposition decrees, the necessity to consider, in the development of monitoring stations, the possibility to detect also data on deposition rates which represent a non-direct exposure of the population through the food chain. For sampling the Directive considers only two types of depositions: for total deposition (bulk and Bergerhoff) and wet only deposition.

Generally, total atmospheric deposition sampling is carried out with cylindrical bottle+funnel devices made of adequate materials (pyrex for organic samples, and polyethylene for inorganic ones); the sample collect in this way made up of dry deposition and humid deposition. With “wet only” deposition samples, the deposition device has a sensor which activates a system which closes or opens the cylinder collector when there is or not precipitation.

The bulk deposition device seems to be the one mainly used, considering the results that it is able to give, its easy employment and moderate costs. By and large, the adopted procedure is the following: depositions are collected by exposing a “cylindrical bottle+funnel” device for a period between a week and a month. The sample collected in the bottle is filtered on a membrane filter, the filter is then mineralized in a microwave absorption system which uses nitric acid and hydrogen peroxide. The acid solution resulting from the mineralization and the acidified filter are analysed by GFAAS or ICP/MS, employing the procedure described by the UNI EN 14902 norm. Two separate samples, collected with two different depositions, are necessary for a simultaneous

determination of PAHs and metals, even if the devices and sampling procedures are substantially the same. For further information, refer to Report ISTISAN 06/38 and the UNI EN norms indicated below [20].

The Italian methods for the determination of arsenic, cadmium, nickel and polycyclic aromatic hydrocarbons in atmospheric depositions [20] is quoted in Legislative Decree 152/2007 (implementing Directive 2004/107/EC) [21] and, after its abrogation, in Legislative Decree 155/2010 (implementing Directive 2008/50/EC) [22]. When the related CEN methods were issued, the European procedures were included in the revision of the air quality regulations (Legislative Decree 24/12/2012, no. 250) [23], and in particular in the following norms:

- UNI EN 15841: 2010; for arsenic, cadmium, lead and nickel [24];
- UNI EN 15853: 2010; for mercury [25];
- UNI EN 15980: 2011; for PAH (benz[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenz[a,h]anthracene and indeno[1,2,3-cd]pyrene) [26].

It should also be mentioned that with Ministerial Directive 29/11/2012 “Identification of special sites for measuring air quality, foreseen by clause 6, sub-clause 1, and clause 8, sub-clauses 6 and 7 of Legislative Decree 13/8/2010, no. 155” four sites have been identified in different Italian Regions (Lombardy, Marche, Puglia, Lazio) [27], to carry out measures of pollutant deposition in particular for: arsenic, cadmium, nickel and mercury, and PAHs of toxicological importance. Three of the sites are in areas which may be classified as “background rural site” and one is in an area which may be classified as “background suburban site”; these will give a consistent amount of data which will be valuable for a national view and as a basis for further in-depth analysis and pattern diffusion studies.

Among the aspects which need further study in that field, there is the finalising of a specific norm for the determination of total atmospheric deposition flux of PCDD/F and PCB. Furthermore, even if there are presently already indications on sampling and methods there still are no specific limits for particulate sediment deposit, nor for its organic and inorganic micropollutant content. In anticipation of the CEN method, the general indications already mentioned for the PAH method may be used, and, for the specific part regarding analysis and preparation, reference may be made to UNI EN norm sections 2, 3, and 4.

ENVIRONMENTAL SURVEYS: PRESENT TENDENCY

The importance to be able to utilize deposition data in the management of environmental surveillance has led, for some Authorisations, to require monitoring campaigns for micropollutant atmospheric deposition rates. For several production (technological) cycles, for example steel plants, foundries, refineries, petrochemical plants, termoelectrical power plants, cement plants, incinerators, to have available deposition data for the surrounding area represent a useful tool for evaluating the polluting burden and the environmental pressure and its evolution. An indication on metals and PCDD/F

Table 4
PCDD/F deposition concentrations in some areas of Italy

Area	PCDD/F (pg I-TE $\mu\text{g m}^{-2} \text{d}^{-1}$)	Reference
Coriano – Rimini (Emilia-Romagna region) (Industrial area – Incineration plants)	0.5-2.9	[38]
Porto Marghera – Venezia (Veneto region) (Industrial area) (Lagoon area)	19.8 28.7	[39]
Taranto (Puglia region) (Industrial area)	0.57-45 WHO-TE $\mu\text{g m}^{-2} \text{d}^{-1}$	[40]
S. Didero – Torino (Piemonte region) (Industrial area – Steel production)	0.212-3.27	[41]
Brescia (Lombardy region) (Industrial area)	1.6-10.9 WHO-TE $\mu\text{g m}^{-2} \text{d}^{-1}$	[42]
Mantova (Lombardia region) (Industrial area) (Background area)	1.20-5.13 1.28-2.71	[28]
S. Nicola di Melfi – Potenza (Basilicata region) (Urban area) (Industrial area) (Background area)	1.76 2.03-2.33 1.47-1.79	[29]

PCDD/F: polychlorinated dibenzofurans.

deposition data, pertaining to campaigns carried out on the national territory in areas characterised by different types of emission sources, are reported in Table 4 [28-30; 38-42]. It should be noticed how, for the stations situated in an urban area, deposition values are greater and, as in the “cold season”, greater PCDD/F values, compared with other areas, have been observed in the remote ones. This confirms that emissions from vehicles and civil heating systems (urban areas) and from biomass combustion for agricultural practices and domestic uses (rural areas) give a not entirely insignificant contribution in terms of micropollutant depositions.

CONCLUSIONS

Atmospheric micropollutant deposition flux measures represent already a widely used strategic element in environmental surveillance and in population exposure evaluation for POPs and metals. With the adoption of the Toxicity Equivalence Factors (TEF), which are periodically implemented and updated [31], risk and exposure evaluations may be carried out in a more valid way.

Deposition monitoring, already in use by government control organizations of various countries, contributes to an important increase in experimental knowledge on pollutant deposition fluxes, on their environmental

fate and on the possible effects on human health. In connection to this, the experiences carried out in Belgium, Germany and France should be highlighted; the results they obtained were used to obtain guide values for PCDD/F and DL-PCB, considering the correlation TDI/deposition [11-14].

Following this trend, already consolidated by various countries, it seems therefore well-timed to promote this measuring method in areas potentially interested in emissions fall-out from plants which use in their technological cycles fossil fuels, biomasses, wastes, or from some steel production processes. Likewise, it is by now necessary to have available guidelines and/or specific limits to better manage particularly difficult situations in the environment and population health which are observed in various national areas. It should be kept in mind that the mere reference to guideline values or to shared scientific knowledge, regarding environmental data evaluation, becomes less effective in managing controls and in decision making.

Conflict of interest statement

No competing financial interests exist.

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