COUPLING OF ATMOSPHERIC MODEL AND MULTIMEDIA MODEL FOR THE EXPOSURE TO HEAVY METAL RELEASED IN THE ATMOSPHERE – APPLICATION TO INDUSTRIAL RELEASES IN A MEDITERRANEAN REGION

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INTRODUCTION

Atmospheric inputs of heavy metals to agriculture systems can be a significant contributor to metal contamination in soil and plants. This problem was identified in several industrial areas (e.g. Vidovic, M. et al, 2005). Because potential health effects of heavy metals on human health are well recognized and because the intake of contaminated foodstuffs (e.g. vegetables, milk) can be the main exposure pathway for humans (e.g. Voutsa, D. et al, 1996), a better understanding of the full-chain exposure system (point source(s)-atmosphere-soil-plant) is essential for future health risk assessments. However, the coupling of atmospheric dispersion models and food-chain multi-media models was scarcely conducted in the past and, often, only for point sources (Glorennec, P. et al, 2005). The originality of our purpose lies in coupling two sophisticated models that respectively simulate the transport of heavy metals in the atmosphere at the European scale (Polair3D model) and the transfer of contaminants in the air-soil-plant-animal system (OURSON model) and applied to a Mediterranean region.

STUDY AREA AND INPUT DATA

Input data used for running the POLAIR3D model are originated from the IREP emission inventory (i.e. French Pollutant Emission Register). Only the emissions of the region under investigation were considered and simulations were also conducted to show the impact of the pollution originated from this region The investigated period is 2001-2004. Meteorological data are provided by the European Center for Medium-Range Weather Forecasts at the following spatial and temporal resolutions: 0.36° x 0.36°, every three hours. Over the 2001-2004 period, POLAIR3D calculated daily averaged heavy metal concentrations in the atmosphere, as well as dry and wet deposition fluxes. The spatial resolution for POLAIR outputs is governed by those of meteorological input data and, as a consequence, grids scale was 50x50 km^2 in our study. Inside each grid, twenty-four land use coverage, corresponding to the US Geological Survey classification, were considered. Dry deposition flux was distributed among these land use coverage types according to their respective presence within the grid.

Outputs from the POLAIR3D model (i.e. dry and wet deposition fluxes) were entered as input data of the multi-media model OURSON. One type of agriculture products were studied in the present study: leaf vegetables (e.g. lettuce) and is assumed to be cultivated and consumed locally.

As a case study, a region situated in the South-East for France and centered around the Martigues city ('Bouches du Rhône' Department) was selected. It is characterized by strong industrialization, with industries potentially releasing cadmium and lead (e.g. chemical, oil refinery, power and steel industries) but also intensive agricultural activity, occupying 41% of the department.

The figure 1 shows the coupling skeleton outline of atmospheric dispersion model POLAIR3D and food-chain multimedia model OURSON. The diagonal represents the principal compartments and the arrows on both sides indicates the various processes of contamination.

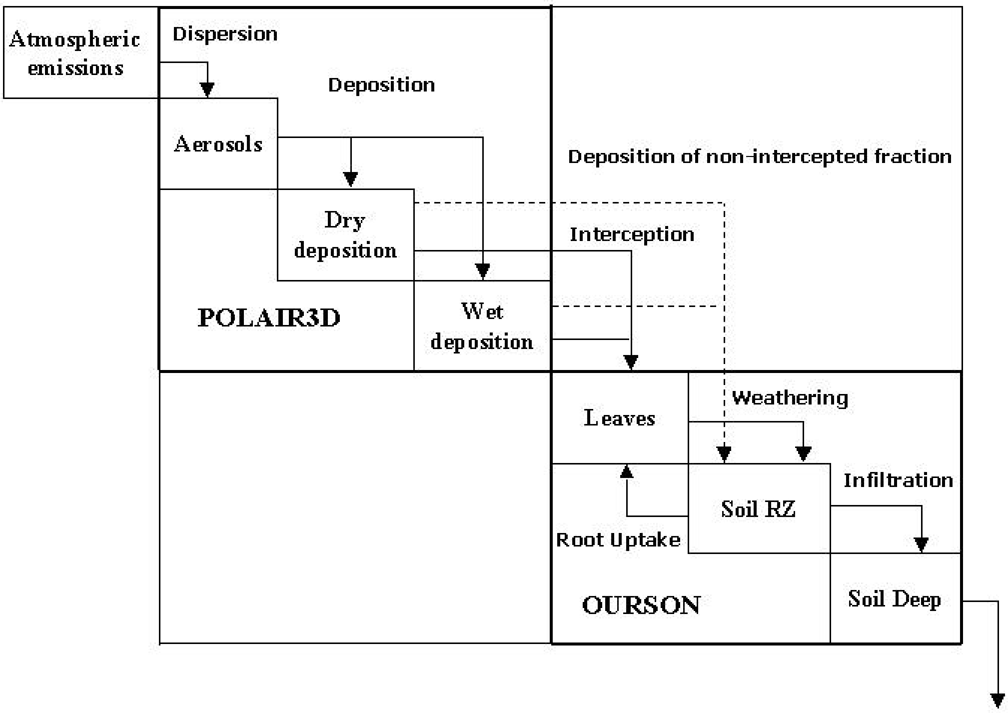

Fig. 1; Features, Events and Processes taken into account in the POLAIR3D and OURSON models.

POLAIR3D MODEL

POLAIR3D is an eulerian three-dimensional chemistry-transport model developed at CEREA (Centre d'Enseignement et de Recherche en Environnement Atmosphérique, http://www.enpc.fr/cerea/fr).

The numerical code is based on a first order time splitting algorithm allowing to separate temporally chemistry (when relevant), advection and diffusion. The advection scheme is a third-order Direct Space Time (DST) scheme. The diffusion scheme is a spatially centered three-point scheme.

Habitually, the heavy metal are considered as belonging to fine particle matter (PM2.5, whose the aerodynamic diameter is lower or equal to 2.5 µm). This comes true for the majority of the compounds generated by anthropogenic sources, which are often processes to high temperature. Versus, the natural processes generate the coarse particle (PM2.5 - PM10).

One of atmospheric pattern consist of allocate to each particle a same median diameter and density representative of population of aerosol.

The POLAIR3D model follows this principle by representing particles as a monodisperse population of aerosol for fixed size.

The processes taken into account in the model are:

- advection/dispersion of aerosols, leading to the calculation of atmospheric cadmium and lead concentration in the atmosphere

- dry deposition of aerosols, the deposition velocity being governed by particles diameters, aerodynamic resistance, quasi-laminar surface boundary layer resistance, surface resistance, gravitational settling velocity, 2D meteorological fields (*Zhang, L. et al.*, 2001)
- wet deposition of aerosols, governed by both in-cloud and below-cloud washout.

For a more detailed description of the model, one can consult *Roustan, Y.* (2005).

OURSON MODEL

Features, events and processes (FEPs) taken into account in the OURSON model are defined in an Interaction Matrix (Figure 1): the main compartments of the biosphere system are identified and listed in the leading diagonal elements (LDEs) of the matrix; the interactions between the LDEs are listed in the off-diagonal elements.

The OURSON compartments and the processes

- Soil surface layer and root zone: Soil surface layer is defined as the soil sub-compartment which is in direct interaction with atmosphere because of dry and wet deposition. Inputs of metals to the soil surface layer are: (1) dry deposition of metals not intercepted by leaves; (2) wet deposition of metals not intercepted by leaves; (3) delayed weathering of metals initially intercepted by plants. Outputs of metals from the soil surface layer is percolation of dissolved metals to the deeper zone (the 'root' zone).
- Plant leaves : Input of cadmium and lead on foliar surface is due to interception of dry and wet deposition of metal by the leaves, while output is due to weathering to the soil surface.
- Edible parts of leaves for leafy vegetables: For the contamination of consumable parts of plants, root uptake and interception on foliar surface are the processes of contamination.

A detailed description of the model is available in *Ciffroy, P. et al* (2006).

RESULTS AND DISCUSSION

Polair3D model

The figures 2 and 3 show atmospheric annual averaged concentration for lead and cadmium obtained from monitoring sites for 2001.

Modelled and measured concentrations in air are generally in good agreement: for lead and cadmium respectively, the ratio between modelled and measured data is lower than 2 in 80% and 75% of cases.

For more than 80% and 75 % (for lead and cadmium, respectively) of points the difference between modelled and measured concentrations in air does not exceed 50 %.

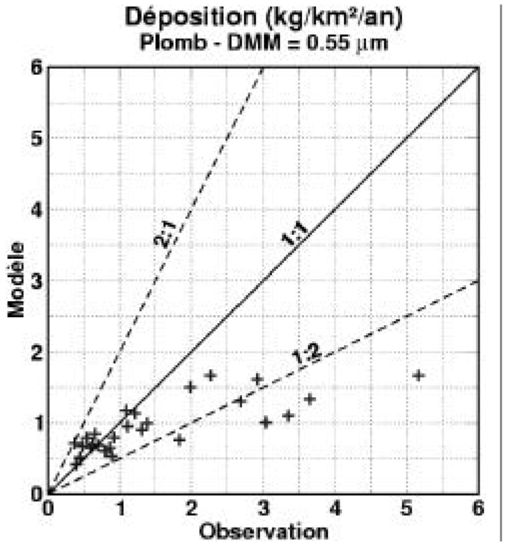

Fig. 2; Comparison of observed and modeled concentrations of lead, in ng/m3, in 2001 (Roustan, Y., 2005).

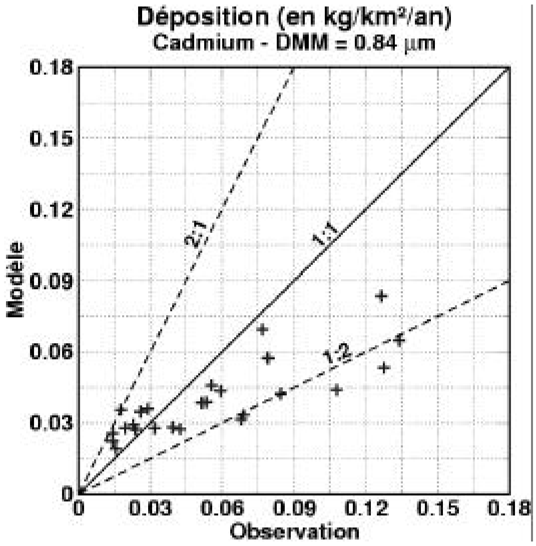

Fig. 3; Comparison of observed and modeled concentrations of cadmium in ng/m3, in 2001 (Roustan, Y., 2005).

OURSON model: Concentration in plants

The concentration in plants was calculated for leaf vegetables. The figure 4 and 5 represent the cadmium and lead concentration in edible parts of leaf vegetable and the wet deposition flux for cadmium and leaf, respectively.

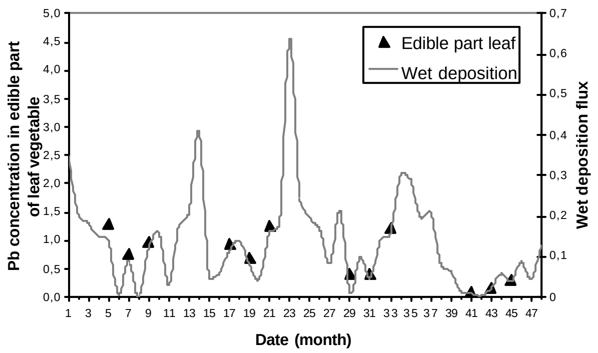

Fig. 4; Lead concentration in edible parts of leaf vegetable (µg/kg DM) and wet deposition flux (µg.m^{-2}.s^{-1}).

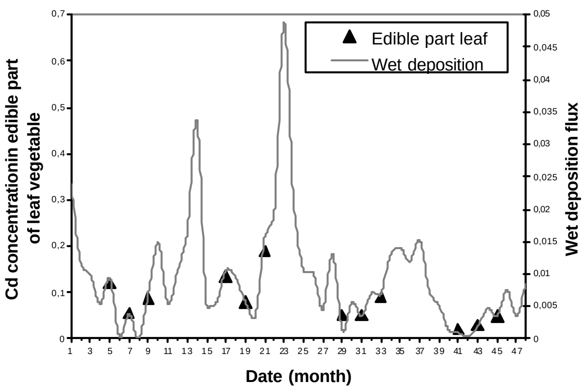

Fig. 5; Cadmium concentration in edible parts of leaf vegetable (µg/kg DM) and wet deposition flux (µg.m^{-2}.s^{-1}).

For the two metal, the concentration in edible parts of leaf vegetable follows the wet deposition flux what seems coherent because the concentration calculation is dependent on the wet flux.

Uncertainty

The objectives of such uncertainty analysis are multiple: (i) because empirical data are very scare, all potential combinations of parameters included in the model must be tested in order to verify that the probability to exceed a given target value is negligible. (ii) determine a confidence interval for the mean annual concentration. For such analysis, each uncertain parameter is described by a probability density function and random samplings were performed on each of these PDFs by Monte Carlo (Latin Hypercube) procedures. When a correlation between two parameters was introduced, random samplings were rearranged according to the Iman-Conover's procedure in order to obtain the desired correlation between the ranks of the correlated parameters. The uncertainty analysis was based on the interpretation of the 5th and 95th percentiles and on the fit to typical distributions (log-normal, log-uniform or log-triangular). (Table 1).

Table 1: Uncertainty analysis for output: concentration in edible part of leaf vegetable.

Output Concentration in edible parts Unit: µg/kg DM	5^{th} percentile	Mean	95^{th} percentile	Ratio $95^{th}/5^{th}$ percentiles	PDF
Cd – leaf vegetable	0.02	0.13	0.47	23.5	log-normal
Pb – leaf vegetable	0.11	0.31	0.98	8.9	log-normal

Besides, the table 2 show the standard and maximal concentration values in leaf vegetable found in the literature for the cadmium and lead, respectively.

Table 2: Standard and values of maximal concentrations in vegetables leaf in the literature for cadmium and lead.

Source	Type de légumes	Concentration
CE 466/2001	Leaf vegetable	200 µg/kg WM for Cd 300 µg/kg DM for Pb
Codex	Leaf vegetable	200 µg/kg DM for Cd 300 µg/kg DM for Pb

The outputs for the best estimation and the uncertainty are compared with the standard and show that no risk is expected when only local releases are considered.

A future study will also assess the contribution of large-scale contamination inputs at Europe scale.

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