Air dispersion modelling for the evaluation of population exposure to pollutants emitted by complex areal sources

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Abstract In this work an application of the CAREA model is presented for the evaluation of the population exposure to pesticides emitted by agricultural fields. CAREA is a Gaussian air dispersion model based on a simplification of the AERMOD formulation because AERMOD is hardly applicable on an high number of complex polygons.

CAREA was applied to pesticides emitted by 1519 agricultural fields and considering 2584 receptors distributed on an area of 8430 km^2. CAREA was run with an hourly time step from March to September. CAREA output provided, for each receptor, a relative concentration value that was assumed proportional to the receptor exposure.

The analysis of the results showed a smooth exposure distribution with 46% of non-exposed receptors instead of the cut off distribution achieved by proximity models. Finally, an experimental measurement campaign was carried out in order to find suitable distances for the assessment of population exposure. To this aim, the air concentration of a pesticide was evaluated by an high volume sampler equipped by quartz fibre filters for aerosols. An AERMOD simulation was performed in order to assess the spatial distribution of the concentration over a test site, thus an experiment was carried out in order to assess the differences between the two models.

Keywords: CAREA, pesticides, Gaussian Air Dispersion Model, Complex areal sources, exposure

1. Introduction

Dispersion models have been widely used to assess the dispersion of pesticides, both relying on preexisting models (Johnson *et al*., 1999) or by the application of specifically developed models, such as AgDRIFT™ (Teske *et al*., 2002), AGDISP™ (Bilanin *et al*., 1989), PERFUM (Reiss and Griffin, 2006). However, these models are usually designed to work with a very limited number of risk sources in micro scale simulations. When thousands of sources need to be considered, traditional models are hardly to be applied due to their computational time and amount of resources. In this work is presented an application of the CAREA model (Complex AREAl atmospheric dispersion model) for the evaluation of the population exposure to pesticides emitted by agricultural fields. The CAREA model, based on a simplification of the formulation of the well-known AERMOD (US-EPA, 2015), provides for each receptor a quantity directly related to its exposure to agricultural land use categories, assumed as sources of pesticides.

This work is part of an epidemiological study aimed at the correlation between ALS (Amyotrophic Lateral Sclerosis) and the exposure to pesticides. Firstly, the exposure of receptors was evaluated by a proximity analysis, approach preferred by epidemiologists for its simplicity and immediacy. Thus, it was wondered whether the effect of meteorology could affect the exposure assessment. For this purpose, CAREA was applied to pesticides emitted by 1519 agricultural fields and considering 2584 receptors distributed on an area of 8430 km^2 including the three provinces of Modena, Reggio Emilia and Parma located in the centre Po Valley.

Finally, an experimental measurement campaign was carried out in order to find suitable distances for the assessment of population exposure. To this aim, the air concentration of a pesticide was evaluated using an high volume sampler equipped by quartz fibre filters for aerosols. Moreover, an AERMOD simulation was performed in order to assess the spatial distribution of the concentration over a test site, thus an experiment was carried out in order to assess the differences between the two models.

2. Methods

Geographical context and dataset

The study area is located in the Po Valley, and includes the provinces of Modena, Reggio Emilia and Parma and covers an area of about 8500 km^2. A dataset of 2584 case/control people were used as target receptors for the assessment of the exposure to pesticides. Receptors are spatially distributed over the three provinces, and constitute a reference population for the study.

The CORINE Land Cover (CLC) map (Bossard *et al*., 2000) was used to identify and extract the agricultural lands. The land cover map used in this work is contained in the land use database of the Geoportal of the Emilia Romagna Region. The land use map of the year 2003 was chosen for the study because temporally overlapped with the case/control dataset. Thus, 1519 agricultural fields of orchards were extracted from the map. At the moment, only orchards were considered as risk sources due to large computational time and amount of resources required by the use of atmospheric dispersion models for epidemiological studies, instead of classics geostatistical models.

The application of the CAREA model

The CAREA model consists in the use of a Gaussian dispersion formulation to simulate mean concentrations and total dry deposition fluxes due to emissions from source areas over flat or undulating zones. CAREA is designed to consider a large number of complex polygons and receptors.

CAREA was applied to 2584 receptor and 1519 complex areal sources of orchards extracted from the land use map of the three provinces of Modena, Reggio Emilia and Parma. The model required as input the coordinates of receptors given by a shapefile where the receptors were listed according to a progressive identifier and with their own coordinates as attribute. Polygons of orchards were provided through another shapefile where each polygon is characterized by a progressive polygon identifier, the code referred to the category of land use of the polygon (in this case only orchards) and the source emission rate. Here, a source emission rate of 0.001 [g/(s m^2)] was used as default value for all the sources considered to achieve a first estimation of exposure of receptors. For the simulation no dry or wet depletion of substances was considered as well as no reactivity or decay of substances was expected. Therefore, we chose an hourly time step from March to September as simulation period relying on meteorological data provided by ARPA Emilia-Romagna and the Osservatorio Geofisico of the University of Modena and Reggio Emilia.

This period of time was chosen to simulate the treatments on orchards (apple orchard, pear orchard, peaches) performed normally from March to September. However, these treatments are not made every day. Consequently, if a constant emission rate is assigned to each sources the exposure of the receptors may be overestimated. But the re-suspension of the pesticide at a time after its application is a phenomenon that needs to be considered (Thatcher and Layton, 1995). Thus, a longer period of time for the simulation can be suitable for the modeling of both phenomena.

In site measurement campaign

A first assessment of pesticide atmospheric concentrations was obtained through an experimental in site measurement campaign. In particular, it was wondered if the use of a simple proximity analysis that considers the exposure constant in all directions is suitable to assess the exposure of population at large scale or the effects of meteorology of the CAREA model must be taken into account for a better evaluation of the receptor exposure.

The measurement campaign involved the use of a high volume sampler during the time of application of an insecticide (Emamectin Benzoate) on the parcels of orchards.

The field measurement campaign was carried out in three days, the 30th of June, the 1st and 14th of July 2014. During the first two days, the air background concentration of the pesticide was detected. To this aim the sampler was placed away from any obstacle in order to work in undisturbed conditions and left switched on for 24 hours. During the second day the air sampling was conducted during the application of a pesticide on orchards parcels. Here, the air-flow was placed downwind at variable distances from the atomizer from a minimum distance greater than 100 m to a maximum distance of about 300 m. The sampling was carried out for the entire duration of treatment in a time period of about 4 hours from 11:00 a.m. to 15:00 p.m of 14th July 2014.

The two fiber quartz filters were analyzed in order to obtain the mass of Emamectin Benzoate sampled. An ultrasonic cleaning was used to extract the substance from the filters. Subsequently, the extract was analyzed in liquid chromatography coupled with mass spectrometry (LCMS / MS) as described in literature by Vega *et al*. (2005).

Numerical simulations

An AERMOD simulation was performed in order to assess the spatial distribution of the concentration over the test site.

The simulation was setup in order to consider both the spray drift at the time of application and the re-suspension of the pesticide at a time after its application. The latter phenomenon was modeled assuming fields as emission sources even after the application of the pesticides. To this aim, the simulation was carried out in four separate runs, because four fields (one of pear orchards, two of pear and apple orchards, one of stone orchards) were involved in the application of the pesticide during the experiment. The pesticide was applied clockwise starting from the upper left pear orchards field (source 1) up to the southernmost pear orchards field (source 4). Therefore, the first run involved only the contribution of the source 1, while the second run involved the contribution of the source 1 and 2. Similarly, the last run involved the contribution of all the sources. The simulation was setup considering a computational domain of 2.5 x 2.5 km^2 focused on the farm. Moreover, a source emission rate of 0.001 [g/(s m^2)] was used as default value for all the sources. Additional simulation parameters involved the use of a simulation period of four hours from 11:00 a.m to 15:00 p.m. of July 14, 2014. For simplicity, no dry or wet depletion of substances was considered as well as no reactivity or decay of substances was expected.

Finally, in order to compare CAREA and AERMOD simulations an experiment over a rural area of 5 x 5 km^2 was considered. Three farm fields were assumed as source of pollutants and a regular grid of 10000 receptors, with steps of 50 m, was defined around the sources. Thus, a generic pollutant for which deposition is negligible was considered. The emission rates of the three sources were set to the arbitrary value of 0.1 [g/(s m^2)]. A preexisting meteorological file was used for the experiment. The simulations were run for February 1990 and July 1990 in order to consider a wide range of instability and stability events.

3. Results and discussion

Application of the CAREA model

The CAREA output provided, for each receptor, a relative concentration value (C) assumed proportional to the receptor exposure to orchards and the concentration ratio (C_{ratio}), that is a normalization of the concentration of the receptor according to a standard case. The CAREA model was compared with a simple proximity model already adopted for the evaluation of the exposure of receptors to pesticides in the current study. The proximity model involved the use of a fixed distance (buffer of 100 meters) between source and receptor as a cut-off threshold to evaluate the percentage of exposure of the receptor itself. In order to evaluate the differences in terms of exposure of receptors, exposure data calculated by the two models were normalized and thus, split and encoded into five risk classes: the code "-999" was assigned to not exposed receptors, while the classes "0, 1, 2, 3" was used to encode normalized exposures between the range [0, 0.25], [0.25, 0.5], [0.5, 0.75], [0.75, 1.00]" respectively.

Observing Table 1, it can be stated that the use of the CAREA model considerably reduces the number of non-exposed receptors, from the 96% of the proximity model to the 46% of the CAREA model. This is a consequence due to the formulation of the methods with a short computing distance of 100 m for the proximity model and the use of a Gaussian model able to simulate the dispersion of pollutant up to few kilometers for the CAREA model. Moreover, by observing Table 1, it can be stated that the differences between the two methods become thinner at high exposures. Especially, when exposure classes "2" and "3" are considered exposures provided by the method are very similar This latter aspect leads to a greater protection for the receptors, because at high exposure levels the output provided by the three levels is less dependent to the chosen approach.

In site measurement campaign

The experimental measurement campaign was organized to achieve a first assessment of the pesticide atmospheric concentrations.

The two fiber quartz filters were analyzed in order to obtain the mass of Emamectin Benzoate sampled during the white measurement and during the application of the pesticide. The analysis of the white filter reported no mass of Emamectin Benzoate within the filter used to evaluate the background atmospheric concentration. Contrariwise, a mass of 0.97 ± 0.31 µg/ buffer was found within the filter used during the pesticide sample. Assuming a flow rate of about 50 ft/min of the TSP high volume air sampler, the calculated atmospheric concentration of Emamectin Benzoate is of 0.02 ng/m^3.

To date, it's not possible to state if the retrieved quantity is consistent or not with the data found in literature because there are too many uncertainties in the factors involved. However, the retrieved quantity allowed to state that distances up to 100-200 m used in the proximity analysis were certainly suitable for the assessment of the exposure of population to pesticides because the concentration of the substance was effectively found within these distances.

Numerical simulations

An AERMOD simulation was performed to assess the spatial distribution of the concentration over the test site. Each simulation provided an output text file where concentration values were given for each grid node: four files were obtained, one for each simulation. Thus, the output files were sum and displayed in a GIS environment to obtain the concentration map over the farm domain (Figure 1). By observing Figure 1, it is possible to state that the dispersion of the pesticide is strongly affected by meteorological conditions with a well developed plume along the main wind direction. Moreover, two transects were extracted along the N-S and E-W directions centered on the sampler.
By observing the N-S transect on Figure 2 it is reasonable to affirm that the concentration value retrieved by the sampler was underestimated along this direction because the concentration profile shows two peaks probably due to the two sources (fields) present north and south from the sampler itself. Contrariwise, the concentration appears correctly estimated along the E-W direction. Moreover, significant concentration value were simulated by AERMOD far from the sampler, especially along the mean wind direction. This is true verified by observing the E-W transect that showed positive concentrations up to 130 meters from the sampler. Once again, this means that the use of a distance of 100 m can be suitable for the assessment of the exposure of population to pesticides.

However, the measurement campaign, as described so far was only a preliminary experiment able to simulate the exposure of receptors at small scale. Thus, if a wider area it is considered, it is reasonable to expect a largest exposure of receptors. Additionally, the dispersion map obtained by the AERMOD simulation showed no-zero concentrations up to distance of 500 m from the treated fields. For these reasons, the use of the CAREA model, that consider the effects of meteorology in the calculation of the exposure, should also be considered a valid approach to calculate the exposure of the receptors to pesticides.

Finally, in order to compare CAREA and AERMOD simulations an experiment over a rural area of 5 x 5 km^2 was considered. Figure 3 shows the scatter plots of ground concentration values achieved by the CAREA and AERMOD models. All the scattered points lie close to the identity line. Thus, from the comparison it can be noticed that CAREA simulations are very close to the AERMOD simulations. Furthermore, by observing the scatter plot of July, it is to note that the CAREA model slightly underestimates AERMOD. This behavior is probably due to the simplification used in the CAREA model that led to underestimation of ground concentrations in unstable conditions.

4. Conclusions

The use of the CAREA model considerably reduces the number of non-exposed receptors, from the 96% of the proximity model to the 46% of the CAREA model. This is a consequence due to the formulation of the methods with a short computing distance of 100 m for the proximity model and the use of an atmospheric dispersion model able to simulate the dispersion of pollutant up to few kilometers for the CAREA model. Moreover, it can be observed that the differences between the two methods become thinner at high exposures. This latter aspect leads to a greater protection for the receptors, because at high exposure levels the output provided by the two methods is less dependent to the chosen approach.

Concerning the experimental measurement campaign a significant concentration value of Emamectin Benzoate was sampled during the application of the pesticide on orchards. To date, it's not possible to state if the retrieved quantity is consistent or not with the data found in literature because there are too many uncertainties in the factors involved. However, the retrieved quantity allowed to state that distances up to 100-200 m used in the proximity analysis were certainly suitable for the assessment of the exposure of population to pesticides because the concentration of the substance was effectively found within these distances.

Moreover, concentration values simulated by AERMOD were also retrieved far from the sampler, especially along the mean wind direction. This is true verified by observing the E-W transect centered on the sampler that showed positive concentrations up to 130 meters from the sampler. Once again, this means that the use of a distance of 100 m can be suitable for the assessment of the exposure of population to pesticides. However, the measurement campaign described so far was an only a preliminary experiment able to simulate the exposure of receptors at small scale. Thus, if a wider area it is considered, it is reasonable to expect a largest exposure of receptors. Additionally, the dispersion map obtained by the AERMOD simulation showed no-zero concentrations up to distance of 500 m from the treated fields. For these reasons, the use of the CAREA model, that consider the effects of meteorology in the calculation of the exposure, should also be considered valid approaches to calculate the exposure of the receptors to pesticides.

The results of the comparison between AERMOD and CAREA models show that CAREA simulations are very close to the AERMOD simulations. Furthermore, by observing the scatter plot of July, it is to note that the CAREA model slightly underestimates AERMOD. This behavior is probably due to the simplification used in the CAREA model that led to underestimation of ground concentrations in unstable conditions.

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Table 1. Number and percentage of receptors characterized by a certain exposure level for each method.

Proximity analysis			CAREA model		
Code	Number	%	Code	Number	%
-999	2469	96	-999	1181	46
0	73	3	0	1233	48
1	25	1	1	153	6
2	12	0	2	10	0
3	5	0	3	7	0
Sum	2584	100	Sum	2584	100

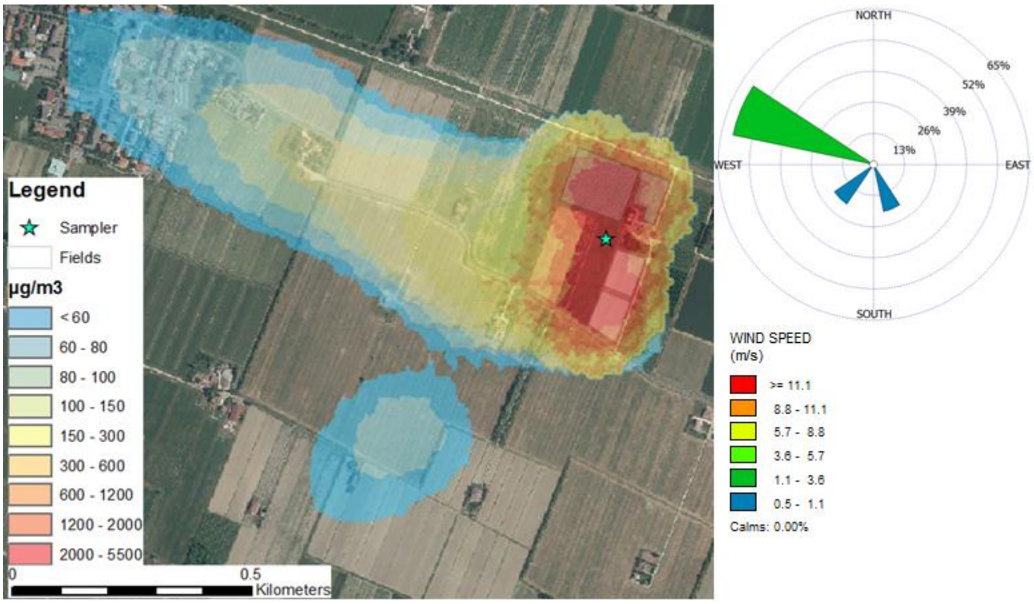

Figure 1. Dispersion map of the pesticide over the test site expressed in terms of relative concentration values. The map only allows a qualitative evaluation of the dispersion of the Emamectin Benzoate since AERMOD was not calibrated so far.

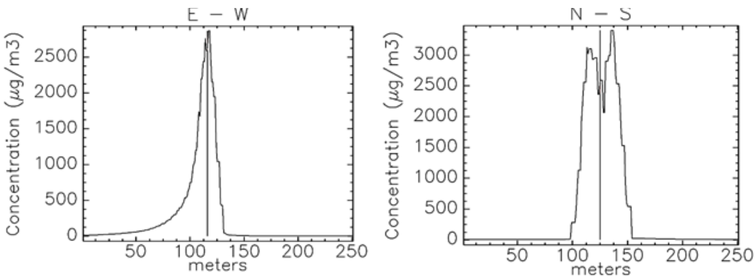

Figure 2. Concentration profiles of the pesticide extracted in the East-West (letf) and North-South (right) direction centered on the sampler. The black line indicates the sampler position.

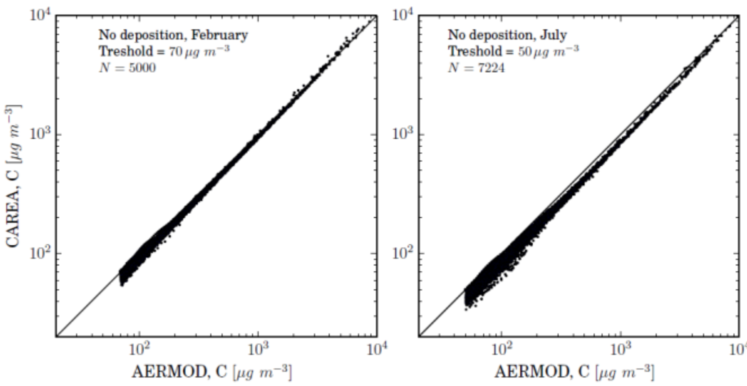

Figure 3. Comparisons (scatter plots) between AERMOD and CAREA concentrations (C) at the receptors (N) with concentrations (AERMOD and/or CAREA) greater than a significant threshold.