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SENSITIVITY OF MODELLED URBAN BACKGROUND OZONE CONCENTRATIONS TO UNCERTAINTIES IN THE GRS INPUT VARIABLES

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Abstract: In this work, we apply the Monte Carlo analysis to evaluate the uncertainty of modelled summer maximum ozone diurnal peak concentrations (C_{max}) in the Metropolitan Area of Buenos Aires (MABA), Argentina resulting from uncertainties in the Generic Reaction Set (GRS) input variables, using the DAUMOD-GRS model. Values of C_{max} occurring at early morning or late evening hours present greater uncertainties than those occurring around midday hours. Uncertainty of C_{max} is dominated by that in the GRS ozone initial concentration at all analysed receptors, with relative contributions varying between 67.5-89.8%. The second most important variable is the nitrogen oxides initial concentration, whose relative contribution may increase (in our experiments up to 11.7%) depending on the uncertainties of the GRS input variables.

Key words: *DAUMOD-GRS, Monte Carlo method, ozone, sensitivity, uncertainty.*

INTRODUCTION

The Generic Reaction Set (GRS) (Azzi et al., 1992) is a simplified photochemical scheme which represents the interactions between nitrogen oxides (NO_x), reactive organic compounds (ROC) and ozone (O_3). It is included in the algorithms of several air quality models (e.g., ADMS-Urban, TAPM, SOMS) and despite of its simplicity it has proved a good ability to simulate ground-level O_3 concentrations at urban scale (e.g., Hurley et al., 2005; Kim et al., 2005). The DAUMOD-GRS model (Pineda Rojas and Venegas, 2013a) has also shown an acceptable performance when tested against concentrations observed at the Metropolitan Area of Buenos Aires (MABA), Argentina (Pineda Rojas, 2014). In a recent work (Pineda Rojas et al., 2016), an uncertainty analysis of modelled summer maximum O_3 peak 1 h-concentrations (C_{max}) due to uncertainties in the model input variables showed that despite being mostly influence by that in the regional background O_3 concentration (which is well known to be a key input variable for the GRS), other variables can also make important contributions. In order to better understand these results, in this work we assess the uncertainty of C_{max} that is introduced by uncertainties in the GRS input variables, and their relative contributions. Since errors in such variables are not really known, the sensitivity of C_{max} uncertainty to them is also analysed.

METHODOLOGY

The DAUMOD-GRS model allows the estimation of ground-level urban background O_3 concentrations, resulting from area source emissions of NO_x and ROC. In the coupling, the DAUMOD model (Mazzeo and Venegas, 1991) feeds the GRS. A detailed description of the coupled model can be found in Pineda Rojas and Venegas (2013a).

The GRS input variables that can be affected by errors in the model input variables are: the reaction constant rates (k_1-k_4) which depend on the air temperature and the solar radiation; the initial concentrations of nitrogen oxides (C_iNO_x) and reactive organic compounds (C_iROC), that are controlled by their respective emissions and the atmospheric transport and dispersion; and the initial concentration of ozone (C_iO_3) which depends on the regional background O_3 concentration and on the remaining concentration from the hour before (i.e., the "memory effect").

In order to evaluate the uncertainty of C_{max} that is introduced by the uncertainties in these seven variables, the Monte Carlo (MC) analysis (Moore and Londergan, 2001; Hanna et al., 2007) is applied. Since the probability density functions for such internal variables are not known, we design three experiments considering log-normal distributions for all variables and different combinations of their possible errors (see Table 1) which are based on values published in the literature (e.g., Hanna et al., 1998). Simple random sampling (Moore and Londergan, 2001) is used to obtain sets of N=100 perturbations for each variable and experiment. The relatively simple coupling between the DAUMOD and the GRS models allows a few code modifications so that the GRS input variables can be perturbed during each simulation. The MC runs are performed for eight selected receptors shown in Figure 1 (see Pineda Rojas et al., 2016), obtaining a set of 100 possible results of C_{max}, from which uncertainty is estimated. On the other hand, these results are also used to perform Multiple Linear Regression Analysis in order to assess the relative contribution from each variable to the estimated C_{max} uncertainty.

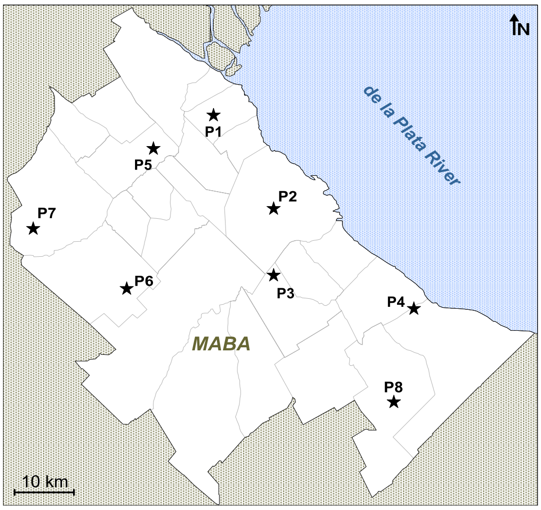

Figure 1. The Metropolitan Area of Buenos Aires (MABA) and selected receptors presenting different emission and atmospheric conditions at the time of occurrence of C_{max}(ZU).

The model input data for the zero-uncertainty (ZU) simulation conditions are the following: surface hourly meteorological information registered at the domestic airport during the 2007 summer, sounding data from the station located at the international airport, high-resolution (1 km^2, 1 h) area source emissions of nitrogen oxides and volatile organic compounds from the emissions inventory developed for the MABA (Venegas et al., 2011), and clean air regional background concentrations for all species (see Pineda Rojas, 2014). The performance of the DAUMOD-GRS to simulate peak O_3 concentrations in the MABA under such conditions has been discussed in Pineda Rojas and Venegas (2013b).

Table 1. Considered errors for the GRS input variables in each experiment

Input variable	Exp-1	Exp-2	Exp-3
k_1-k_4	30%	30%	30%
C_iNO_x	30%	50%	80%
C_iROC	30%	50%	80%
C_iO_3	30%	30%	40%

In this work, at each of the eight selected receptors, the uncertainty of C_{max} (taken as the 95% confidence interval) is approximated by that of O_3 hourly concentration at the time (day of summer and hour) of occurrence of C_{max}(ZU).

RESULTS

The eight selected receptors (Figure 1) present a wide range of atmospheric and emission conditions at the time of occurrence of C_{max}(ZU), with varying wind speed and direction, air temperature, sky cover, atmospheric stability class, solar radiation, and NO_x and ROC emission rates. The order of the receptors is in the direction of decreasing emission rates. The wide range of atmospheric conditions is related to the varying hour of occurrence of C_{max}(ZU): around midday hours at receptors P1-P5, and in the early morning or late evening at receptors P6-P8. C_{max}(ZU) values simulated at these receptors are in the range 16.3-26.2 ppb (see Pineda Rojas et al., 2016).

Variation of C_{max} uncertainty among selected receptors

In order to analyse the C_{max} uncertainty under such different environmental conditions, we first consider the results obtained in the Experiment 2 (Exp-2) whose uncertainties values are commonly found in the literature (e.g., Hanna et al., 1998) and because they represent a kind of average between the three experiments.

Mean C_{max} values obtained from the MC runs under conditions of Exp-2 vary between 16.4-26.1 ppb and tend to those of C_{max}(ZU), indicating that convergence is achieved for N=100 simulations. On the other hand, the coefficient of variation (σx100/mean) of C_{max} varies between 14.5-28.5%, being consistent with results obtained in other places (e.g., Bergin et al., 1999; Hanna et al., 2005).

Figure 2 shows the 95% confidence range of C_{max} obtained at each receptor under conditions of Exp-2. A great difference of the uncertainty is observed between receptors P1-P5 (10.6-12.1 ppb) and P6-P8 (19.0-22.2 ppb), which shows that the uncertainty of C_{max} at receptors where the O_3 peaks occur during early morning or late evening hours is almost twice that obtained at receptors where the peaks present around midday hours.

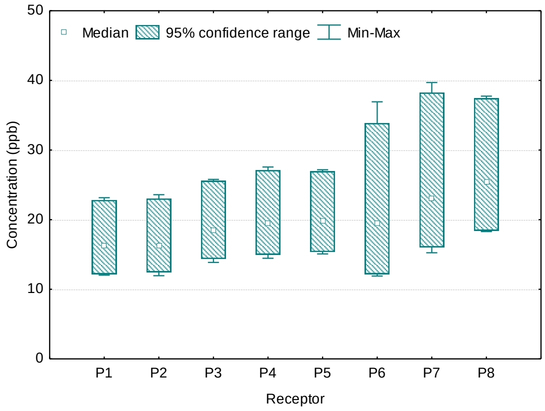

Figure 2. Uncertainty of C_{max} at each selected receptor, under conditions of Experiment 2

At all receptors, the uncertainty in C_iO_3 is the main contributing source to C_{max} uncertainty (67.5-89.8%); the second most important variable is C_iNO_x (0.2-5.0%); while the reaction constant rates k_1-k_4 (k's) represents the lowest contribution (up to 3.1%) (see Table 2). By comparing these contributions in ppb, it is observed that that of C_iO_3 varies between 7.7 ppb (at P1 and P2) and 17.4 ppb (at P7).

Table 2. Uncertainty contribution [in % and ppb] of the GRS input variables to C_{max} uncertainty at each receptor, under conditions of Experiment 2. k's denotes the sum of k_1 to k_4.

Receptor	k's		C_iNO_x		C_iO_3	
	%	ppb	%	ppb	%	ppb
P1	3.1	0.3	4.7	0.5	72.7	7.7
P2	2.7	0.3	5.0	0.5	73.2	7.7
P3	0.5	0.1	1.2	0.1	84.7	9.5
P4	0.4	0.1	1.1	0.1	85.5	10.4
P5	0.2	<0.1	0.4	<0.1	88.5	10.2
P6	0.2	0.1	4.2	0.9	67.5	14.6
P7	0.2	<0.1	2.7	0.6	78.6	17.4
P8	<0.1	<0.1	0.2	<0.1	89.8	17.1

Sensitivity of C_{max} uncertainty to the GRS input variables' uncertainties

Figure 3 presents the greatest difference of C_{max} uncertainty obtained among the three experiments. At all receptors, the lowest C_{max} uncertainty is obtained under conditions of Exp-1. The greatest difference of the 95% confidence range at receptors P1 and P2 is found between Exp-1 and Exp-2 (1.0-1.5 ppb). At receptors P3-P8, the greatest differences occur with Exp-3 and vary between 3.1-5.0 ppb.

The comparison of the relative contributions from the uncertainties in C_iNO_x and C_iO_3 to C_{max} uncertainty between the three experiments is shown in Figure 4. The contribution of the k's is not shown because it varies in its second decimal. The contribution from C_iNO_x increases from Exp-1 to Exp-3 as long as its uncertainty does, with a maximum-to-minimum ratio varying between 2.7 (P6) and 3.8 (P8) (i.e., greater than the variation of its uncertainty). The uncertainty contribution of C_iO_3 is lower in Exp-2 and greater in Exp-3, at all receptors. The maximum-to-minimum ratio varies between 1.0 (P1) and 1.2 (P4), indicating that the uncertainty contribution of C_iO_3 is less sensitive to changes in its uncertainty range than that of C_iNO_x.

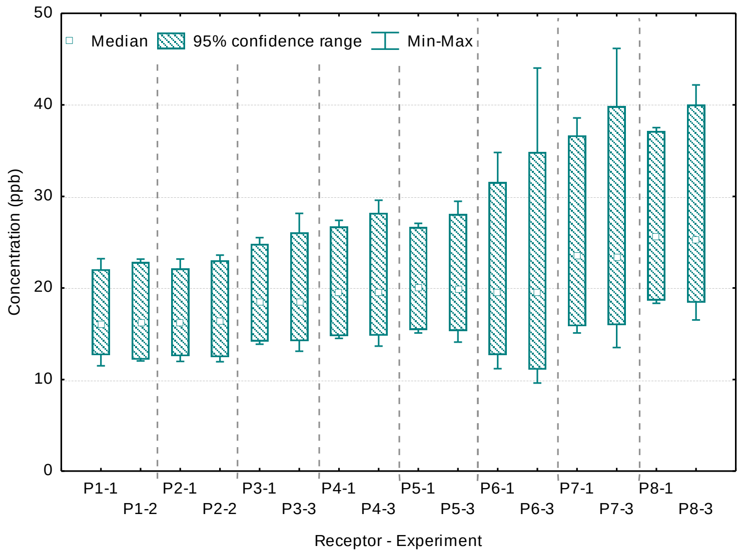

Figure 3. Uncertainty of C_{max} at each selected receptor, under conditions of the two experiments presenting the greatest difference

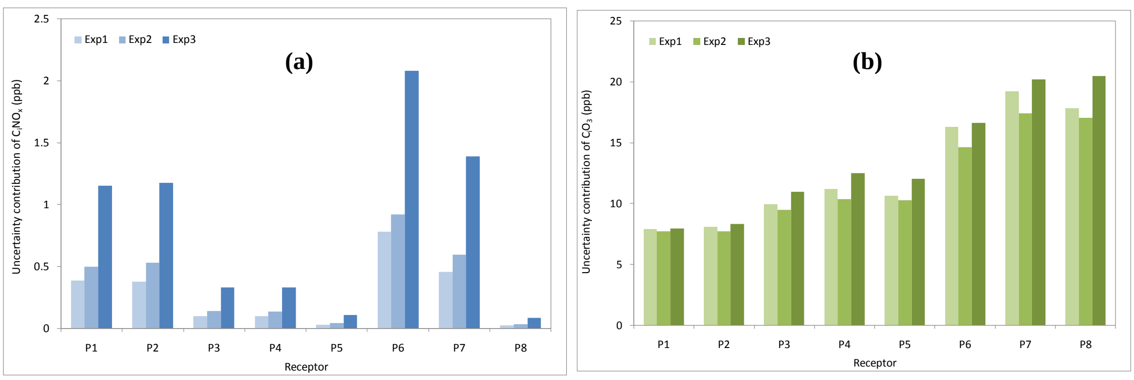

Figure 4. Uncertainty contribution (ppb) of C_iNO_x (a) and C_iO_3 (b) to C_{max} uncertainty at each selected receptor obtained under conditions of experiments 1, 2 and 3

CONCLUSIONS

An uncertainty and sensitivity analysis of the summer maximum O_3 peak concentrations (C_{max}) simulated with the DAUMOD-GRS model to uncertainties in the GRS input variables shows that, under the environmental conditions of the Metropolitan Area of Buenos Aires (MABA):

- Uncertainty of C_{max} (taken as its 95% confidence range) varies spatially, being greater at receptors where C_{max} occurs in the early morning or late evening than where peaks occur around midday hours.
- The relative contributions from the GRS input variables vary spatially also, although the initial concentration of ozone dominates at all analysed receptors.
- The sensitivity of C_{max} uncertainty to the input variables' uncertainty varies among the selected receptors between 1.0 ppb (10%) and 5.0 ppb (26%). The relative contribution of NO_x initial concentration is more sensitive to its uncertainty than that of O_3.

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