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DEVELOPING A FAST PHOTOCHEMICAL CALCULATOR FOR AN INTEGRATED ASSESSMENT MODEL

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Abstract: The use of integrated assessment models, in air quality policy support systems, which combine atmospheric models with others coming from different fields, raises the need of developing specific air quality modeling concepts. Existing air quality models are not directly suitable to integrated assessment models because of flexibility and speed are required in order to provide results to support policy decisions in a reasonable time frame. The air quality model, AYLTP, uses an adapted version of the Lagrangian particle model AUSTAL2000 to calculate the transport and the dispersion along with a coupled photochemical module developed to calculate ozone rapidly using a quasi-first order reaction. Two approaches are compared: one using a look-up table, the other using a coupled box model. The lookup table has been built using the OZIPR model by simulating a large set of possible combinations of meteorological variables and precursor concentrations.

In this approach the ozone concentration variations are obtained using a rate coefficient that is used to affect the mass carried by the Lagrangian particles. The second approach consists of coupling the Lagrangian model AUSTAL2000 with the OZIPR box model, also using a quasi-first order reaction rate. We discuss the differences and the practicability of the two model versions, exploring CPU times, memory requirements, flexibility, applicability, accuracy, implementation and future improvements. The first results of both approaches for a simple case study of Luxembourg shows that using the AUSTAL2000-OZIPR coupled version the CPU is significantly slower. We conclude that the use of tabulated rates is more suitable for this optimization framework albeit it needs further development in accuracy.

Key words: *Air quality, Fast Photochemical Calculator, Ozone, Luxembourg.*

INTRODUCTION

Nature is the most complex example of an integrated system. In practice much simplification is needed to model such systems. As it is impossible to model the whole environment in detail, the sub-systems of a bigger system are isolated. Integrated Assessment models (IAM) isolate the most important sub-systems in order to address integrated solutions towards a given problem, in a world with increasing complexity of questions (Haurie, A. *et al*, 2004).

The Air Quality Integrated Assessment Models form a class of IAM which aims at addressing issues related to atmospheric pollution. They relate, at least, two different realms, energy and air pollution, which are linked by a common component: emissions. Over the last decades important advances in computer technology have contributed to the improvement of air quality models. These models are highly detailed, regarding the atmospheric processes, the interactions between atmosphere and biosphere, and the chemical reactions. However the technology gains are not, yet, sufficient to allow the use of these models within IAM in a reasonable way. 3D-eulerian models are very demanding in CPU time, input data and computational capacity, that can only be provided by high performance computing platforms (Seaman, N.L., 2003). These models are useful to simulate abatement policies scenarios and to compute the subsequent air pollution levels (cascading mode). However 3D-eulerian models are not convenient when the question is posed in the inverse way, that is, find the optimal measures to meet Air Quality (AQ) standards. In this case they are not practical to run under optimization frameworks, in which the sub-models has to iterate until the optimal solution is obtained.

The use of integrated assessment models is not new in AQ policy support. The most known IA models are RAINS/GAINS models developed by IIASA. RAINS (Alcamo, J. and Hordijk, L.,1990) is a major IAM, developed initially to study the acid rains. The following versions extend its capacity to handle other air pollutants as defined by the Convention on LRTAP (Schopp, W., *et al*., 1998), and greenhouse gases as defined in the NEC directive (Klaassen, G., *et al*, 2004). These IAMs are based on EMEP, an Eulerian dispersion model at regional level. The work by (Amann, M., *et al.*, 2001) presents a first attempt of using the RAINS model in optimization mode. Later, the MERLIN model sets up an optimization framework around the RAINS model based on genetic algorithms (Reis, S. Et al., 2005). The EC4MACS model contains the RAINS model along with a set of multidisciplinary models (EC4MACS,2008). The use of such integrated models is increasing and for practical reasons it is important to be able to simplify AQ models, keeping the essential information.

The Luxembourg Energy-Air Quality model (LEAQ), developed by the CRP Henri Tudor, is an air quality integrated assessment model designed for the Grand Duchy of Luxembourg (Zachary, D. S., *et al*., 2011). Figure 1 shows the structure of the model, which is composed of a techno-economic model ETEM and an air quality model AUSTAL2000-AYLTP, linked by an optimization master program that establishes the "dialogue" between the sub-models (Haurie, A. *et al*, 2004).

LEAQ contains an optimization routine which iterates towards an optimal solution. At each iteration, the methodology needs the sensitivity of the AQ model, which is estimated by finite differences. Hence the AQ model is evaluated $(n_v + 1) \times n_i$ times, where n_v, is the number of decision variables (perturbed runs) plus a non perturbed run and n_i is the number of iterations. Therefore the CPU time of the AQ model is crucial to obtain an optimal solution in a reasonable time frame. In this paper we present the AUSTAL2000-AYLTP that was designed for the optimization framework of the LEAQ model.

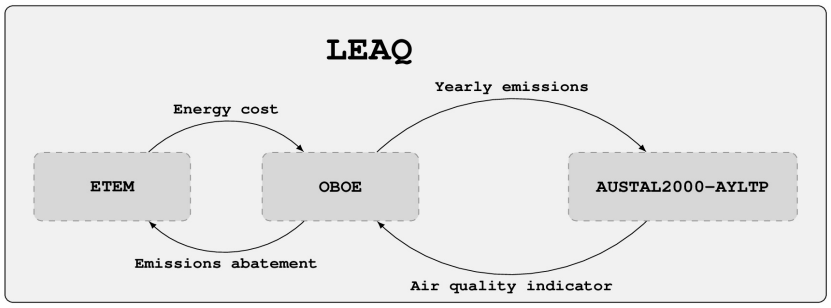

Figure 1: Overview of the IA - LEAQ model, showing the two sub-models and their relation.

THE AIR QUALITY MODEL

AUSTAL2000-AYLTP has been developed from the German Regulation model on air quality control, AUSTAL2000 (Janicke, U. and Janicke, L., 2004). The choice of the model resulted from a survey of the existing open source AQ models (Reis, L.A., *et al.*, 2009). AUSTAL2000 is a Lagrangian particle model, hence it is based on particle trajectories instead of fluxes. This approach avoids the numerical diffusion which is generally associated with the numerical schemes used to solve the partial differential equations intrinsic to the Eulerian models. Furthermore the Lagrangian particle approach adds others advantages, as being a fast transport calculator, allowing source backtracking and having a flexible domain and resolution. The most interesting characteristic of this model is the ability to gain CPU time when the number of Lagrangian particles is reduced however at the cost of loss of accuracy in the results.

On the other hand, in Lagrangian models, the chemistry is decoupled from the transport, and this is one of their main disadvantages. High order chemical reactions are very difficult to implement (Nguyen, K.C., *et al*., 1994), it is nevertheless possible to implement linear transformations (VDI, 2000). Ozone is a pollutant with non-linear behaviour and the implementation of a fast photochemistry module needs groundbreaking solutions.

In this work we use quasi-linear reactions rates to mimic the behaviour of ozone. Despite the fact that ozone has a non-linear behaviour, we can assume that under a certain number of restricted conditions a linear reaction rates holds. A set of variables, which have a high influence on ozone behaviour, was chosen to be the most significant, including zenith angle (θ), relative humidity (RH), temperature (T), NO_x , VOC and O_3 concentrations. We use the box model, OZIPR (Gery, M.W. and Crouse, P.R., 2000), to calculate the reaction rates.

A realistic interval is set for each variable and OZIPR is then run for all the possible combinations between them. The initial conditions are stored in the lookup table (LUT), and the resulting reactions rates Ks are calculated for all the reactive pollutants p according to:

$$Ks_p(c_p(t),T,RH,\theta) = \frac{c_p(t+1,c_p(t),T,RH,\theta) - c_p(t)}{\Delta t},\ p \in \{1,2,3\}, \quad (1)$$

where c is concentration, t is time. The final concentration c p (t + 1, c p (t), T, RH, θ) is calculated by OZIPR and is dependent on the initial concentrations of the pollutants and the meteorological conditions. The initial concentrations are calculated using AUSTAL2000-AYLTP.

The LUT is then plugged with AUSTAL2000, and is called at every cell given a 6 dimension index. The LUT returns the reactions rates Ks. For all pollutants, and at each cell (i, j, k) of the space S = {i ∈ 1, . . . , Nx; j ∈ 1, . . . , Ny; k ∈ 1, . . . , Nz}, where Nx, Ny, Nz, are the number of cells in each direction, these rates Ks are applied as follows:

$$c'_{p,i,j,k}(t) = c_{p,i,j,k}(t) + Ks_p(c_p(t),T,RH,\theta)\Delta t,\ \forall p \in \{1,2,3\};\forall (i,j,k) \in S, \quad (2)$$

where c'p,i, j,k (t) is the concentration resulting from the photochemical reaction. The new concentration is then applied over the particles located in the cell, by distributing the mass equally. Afterwards the Lagragian particles are moved for the next time step carrying the photochemical transformed mass.

This approach allows the simulation of ozone production/depletion using a linear rate, although incorporating the influence of meteorological variables as well as precursor concentrations. It is assumed that the rate is steady in the Δt interval.

Photochemical module versions

For fairness with the developer of AUSTAL2000 (core calculator) we rename the model here as AUSTAL2000-AYLTP, which is the adapted version of AUSTAL2000 coupled with a photochemical module AYLTP.

Figure 2 shows AUSTAL2000-AYLTP model structure, where the dashed lines represent the two approaches applied in this work. The first one is described in detail above where AUSTAL2000 is plugged to the LUT to obtain the reaction rates, and a second one where AUSTAL2000 is plugged directly to OZIPR model. In the latter, the reaction rates are not stored, but instead the current values of the variables are used as initial conditions to run OZIPR and the reactions rates are calculated immediately. In both cases the mass transformation is carried out by using the same equations 1 and 2.

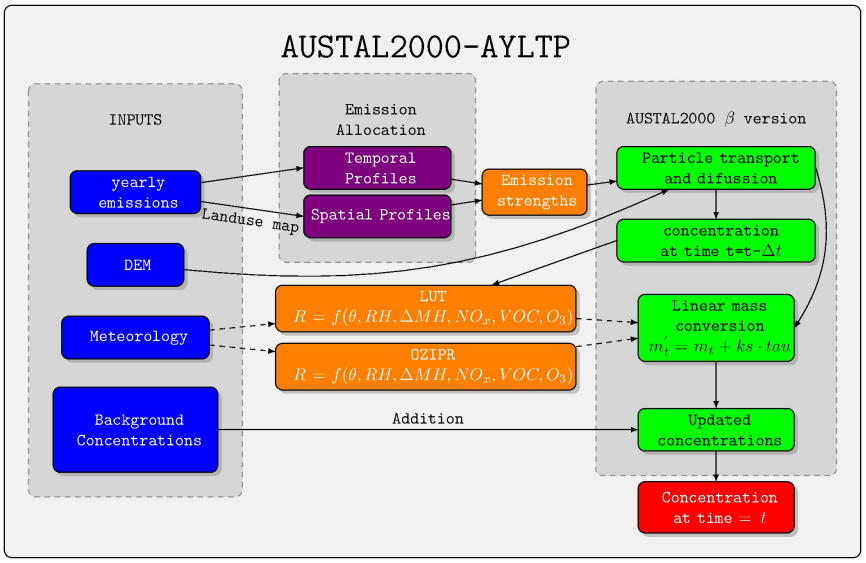

Figure 2. AUSTAL2000-AYLTP Structure, including both versions of the model represented by the dashed line. The Photochemical modules represented by LUT and OZIPR blocks respectively.

Both versions of the model were run for the day 19-07-2006, representing a real ozone episode in the territory of Luxembourg. Only emissions from national territory were considered. The total emissions per economic sector were provided by Long Range Transboundary Air Pollution, the biogenic VOC emissions were provided by the Aristotle University of Thessaloniki. The temporal disaggregation was performed using the information coming from GENEMIS project (Friedrich, R. and Reis, S., 2005), and the spatial allocation was performed by sector using Luxemburg's land use maps. Both the LUT and AUSTAL2000-AYLTP were run with a time step of one hour. The meteorology cal data was provided by the *Administration des Services Techniques de l'Agriculture* (ASTA), the station used is Luxembourg Merl. We present here the comparison of both approaches and the preliminary results of the model.

RESULTS AND DISCUSSION

The results of the methods comparison are presented in Table 1. The LUT version is considerably much faster than the version that runs with OZIPR. All the simulations show that the LUT version is at least 10 times faster than the OZIPR one. The IAM LEAQ requires an answer in a reasonable time range, to be able to find an optimal solution though its iterative process. The use of the LUT showed an important CPU gain, which demonstrates that the CPU time of AUSTAL2000-OZIPR is not compatible with the integration in the LEAQ model.

On the other hand the two versions showed differences in the results. The difference in the averages of the reaction rates and in the averages of the concentrations are presented in the Table 1. These differences increase with the number of cells in the simulation and decrease with the refinement of the time step. For small domains, with high resolution (low number of cells), the differences encountered in the rates do not translate in a different concentration result. However for lower resolutions the differences in the rates have more influence in the average concentration of each cell, since there are less Lagragian particles per cell. The largest differences in the average concentrations are observed when the time step is changed from 10 minutes to one hour. This aspect suggests that the rate variation during one hour is high, and therefore it should be calculated with a higher frequency. Three options are considered regarding the improvement of the LUT: (I) refine the variables interval of the LUT, (II) reduce the time step, so that the rates can be updated more often, (III) the use of an advance krigging method to retrieve the rates values.

The present LUT has 2555280 possible values, which correspond to 10 initial temperatures, 9 relative humidity values, 14 possible zenith angles, 12 ozone initial concentrations, and 13 VOC and NO_x concentrations. The differences in the rates are due to the fact that the LUT is limited in terms of the number of combined initial conditions that can be stored. Therefore the initial conditions are rounded to the nearest value stored in the table.

The change observed in CPU time between the 3rd and 4th simulation is because the simulation of OZIPR for one hour is longer than for 10 minutes time step, even though more time steps are carried out.

The OZIPR version assures no approximation error, and allows for more flexibility because AUSTAL2000 model can hand over to OZIPR input all the possible variables that OZIPR takes (e.g mixing height, not taken into account in this study due to lack of data).

Table 1. Simulation results of both model versions

Number of cells	Time step (min.)	CPU time (min.)		Differences in average	
		LUT	OZIPR	conc. [µgm^{-3}]	rates [µgm^{-3}s^{-1}]
25	10	4	78	0.000	0.0030
120	10	9	96	0.611	0.0017
480	10	11	184	1.643	0.0012
120	60	3	419	15.92	0.4540

The pictures 3 and 4 illustrate the results of the AUSTAL2000-LUT simulation. The simulation was carried out using the national emissions, to which no background concentrations have been added, due to non availability of data. The stations of Mont St. Nicolas, Beckerich and Esch-sur-Alzette show border effects. These effects are explained by the lack of sources at the borders, thus lower concentrations are predicted. The figure 5 shows the comparison with measurements. The differences, are considerable, although the missing background data can explain the differences in magnitude. The model foresees the episode in the last day, but the peak happens later in time. Two factors are likely to explain this event: (I) an increase of the precursors emissions in the afternoon that extended though the evening; (II) in the day 18-07-2006, before the episode there was a peak of NO_x early in the morning (at 9 hours) that the model was not able to foresee, which together with the appropriate meteorological conditions provoked an earlier ozone peak.

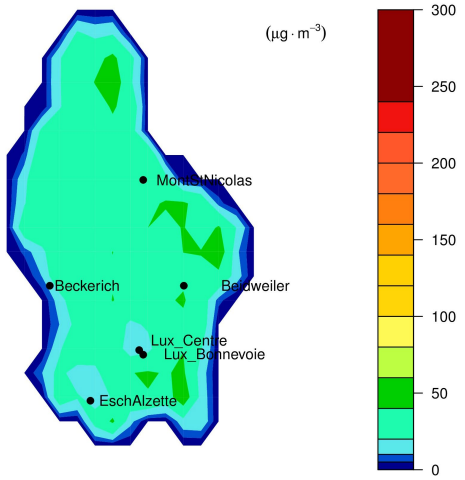

Figure 3. Modeled ozone concentrations (µg.m^{-3}) at 9 hours of 19-07-2006, the points represent the monitoring stations.

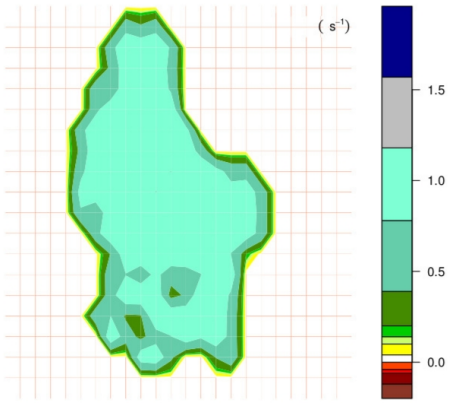

Figure 4. Spatial distribution of the ozone rates(s^{-1}), for the time step corresponding to 9 hours of 19-07-2006.

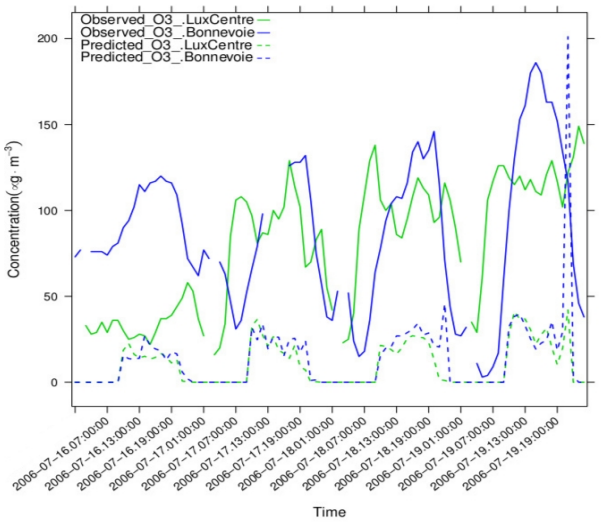

Figure 5. Comparison with the observed ozone values, using the stations of Bonnevoie and Luxembourg centre.

CONCLUSION AND FUTURE DEVELOPMENTS

The work compared two possible methods to obtain the reaction rates used to mimic the tropospheric ozone production. This comparison revealed differences in the reaction rate values. For small domains these differences do not cause variations in the average concentrations. Nevertheless, in larger case studies the differences in the reaction rates cause perturbations in the average concentration, although not significant. The largest difference is observed for the one hour time step simulation. This

fact shows that for a 1 hour time step the use of the LUT is not adequate. The CPU time for the 10 minutes time step are nevertheless still reasonable.

The LUT version offers a more suitable approach for the LEAQ model. The objective was to build a model that could run on approximately 10 minutes. The LUT CPU time is then considered satisfying, for a decision making time frame.

In the LEAQ model, the air quality results are based on the total primary emissions from each techno-economic period of the energy model. Approximations are therefore acceptable. As the ETEM model predicts for such a long term the whole meta-model has inherently large uncertainties associated with long time scales. Consequently, a highly accurately, a CPU costly air quality model would not be desirable in this application.

As a conclusion the gains in CPU time obtained with LUT take advantage over the highest precision obtained with OZIPR at a much higher CPU time. This aspect and the fact that the LUT method presents more potential improvement is the reason for deciding in favour of the use of LUT.

The development of such methodologies is of extreme importance when considering IAM. The use of quasi-linear reaction rates obtained with the help of a LUT, represents an innovative step towards the use of simplified air quality models that involve complex chemistry. This method supplies a good compromise between CPU time and accuracy.
The model AUSTAL2000-AYLTP (LUT version) uses a new approach and requires further testing and validation. It is then essential to incorporate background concentration data in order to perform a proper validation of the model with measurements.

REFERENCES

Haurie, A., Kobler, J.J.E., Clappier, A. and H. van den Bergh. A Metamodeling approach for integrated assessment of air quality policies. Environmental Modeling and Assessment, 1 December 2004.

Seaman, N. L., Future directions of meteorology related to air-quality research. Environment International, (29):245–252, 2003.

Alcamo, J. and Hordijk, L. The RAINS model of acidification: science and strategies in Europe. Dordrecht, The Netherlands: Kluwer Academic Publishers, 1990.

Schopp, W., Amann, M., Cofala, J., Heyes, C. and Klimont, Z. Integrated assessmentof european air pollution emission control strategies. Environmental Modelling and Software, 14(1):1–9, October 1998.

Klaassen, G.,. Amann, M., Berglund, C., Cofala, j., Glund-Isaksson, L. H., Heyes, C., Mechler, R., Tohka, A., Schppp, W. and Winiwarter, W., The extension of the RAINS model to greenhouse gases. Technical Report IR-04-015, International Institute for Applied Systems Analysis, 2004.

Amann, M., Heyes, C., Makowski, M. and W. Schoopp. An optimization model for the control of regional air quality in europe. In J.B.H.J. Linders, editor, Modelling of Environmental Chemical Exposure and Risk, pages 193–203. Kluwer Academic Publishers, The Netherlands, 2001.

Reis, S., Nitter, S. and Friedrich. R., Innovative approaches in integrated assessment modelling of european air pollution control strategies - implications of dealing with multi-pollutant multi-effect problems. Environmental Modelling & Software, 20:1524–1531, 2005.

EC4MACS. Life iii ec4macs european consortium for modelling of air pollution and climate strategies. Technical report, International Institute for and Applied Systems Analysis, June 2007.

Zachary, D.S., Drouet, L., Leopold, U. and Reis, L.A., Trade-offs between energy cost and health impact in a regional coupled energy–air quality model: the LEAQ model. ENVIRONMENTAL RESEARCH LETTERS, 6(024021):9, 21 June 2011.

Janicke, U., and Janicke, L., Lagrangian particle modelling for regulatory purposes - a survey of recent developments in germany. In Proceedings of the 11th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, pages 2000–2004, 2004.

Reis, L.A., Zachary D. S., Leopold U. and Peters B..,AYLTP - a fast air quality calculator. In Air Pollution 2009, 2009.

K.C. Nguyen, J.A. Noonan, and W.L. Physick. Comparison of eulerian and lagrangian model predictions of power station plume dispersion. In Transactionson Ecology and the Environment, volume 3, 1994.

VEREIN DEUTSCHER INGENIEURE. VDI 3945 Part 3; Environmental meteorology; Atmospheric dispersion model; Particle model. Technical report, Verien Deutscher Ingenieure, September 2000.

Gery, M.W. and Crouse, R.R., User's guide for executing ozipr. Technical report, U.S. Environmental Protection Agency, 1990.

Friedrich, R. and Reis, S., Emissions of Air Pollutants - Measurements, Calculations and Uncertainties. Springer.