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Interactive comment on "Sensitivity analysis by the adjoint chemistry transport model DRAIS for an episode in the Berlin ozone (BERLIOZ) experiment" *by* K. Nester and H.-J. Panitz

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We thank the referees for their helpful comments on our paper. We will try to take into account the suggestions and recommendations in the revised version as far as possible. In the following we discuss the comments of both referees separately.

Discussion of the comments of referee #1:

Referee #1 has divided his comments into two blocks.

The first part deals with the quality of the model KAMM/DRAIS. As the expression "model" already implies, a model cannot be perfect. The basic approximations in the

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KAMM/DRAIS model are realistic enough in order to calculate the relevant meteorological and chemical quantities in mesoscale areas. Several evaluation studies, in which our model system was involved, have shown that the results are as good as those from comparable models or even better. The objection of the referee that the parameterizations used in the model are poorly described is accepted by us. We will go into more details in the revised version. The behaviour of our model concerning NOx and VOC limited regimes in the city plume of Berlin is discussed in the paper by Hammer et al. (2002). They show that both stations Eichstädt and Menz are located in the VOC limited area.

In the second part of his review referee # 1 criticizes that we did not consider meteorological parameters in our sensitivity study. He mentions the boundary layer height as one important parameter.

We put much effort into the modelling of the meteorological conditions. In order to get a good agreement between the measured and simulated meteorological quantities, several simulations with the meteorological model KAMM have been carried out using a nudging procedure. In the paper we restrict the examples to the comparison of wind direction between model results and aircraft measurements. It demonstrates the good quality of the meteorological modelling, and that the model even succeeded to simulate the strong wind direction change occurring above the boundary layer (Fig. 4 in our paper). Further comparisons of the wind speed, the temperature, and the humidity support the good agreement between modelled and measured meteorological quantities.

The boundary layer height has the greatest effect on the profiles of the species concentrations in the source region. In our case this is the city of Berlin. With increasing distance from the sources, errors in the estimation of the boundary layer height are less important with respect to ozone concentration profiles, especially in the afternoon. The ozone concentrations over the city of Berlin and at the station Eichstädt (30 km downwind of the city centre) are calculated realistically and it can be concluded that

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the boundary layer heights in these areas are well simulated. Therefore, it is rather unlikely that such a great error in the modelled boundary layer height in the area of Menz occurs that the observed discrepancies in the ozone concentrations at this station can be explained. In summary, the simulation of the meteorological conditions is realistic enough that it is not necessary to consider meteorological parameters in the sensitivity study.

Another fact supports our statement. The BERLIOZ episode we are considering has also been modelled by the EURAD group, University of Cologne, using their EURAD model. This model uses the same chemical mechanism as the DRAIS model. But the parameterizations in the meteorological part are different from those in the KAMM model. However, the results of the EURAD model show similar ozone concentrations at the station Menz as the results of our model. Therefore, we conclude that this is an additional indication that the deficit we are looking for is caused by the chemistry model.

The referee mentions the splitting of the vertical atmospheric column into four layers only. These four vertical layers are related only to the mass balance calculation, and not directly to the sensitivity study. The mass balance calculation was the issue of a previous study cited in the paper (Panitz and Nester, 2002) We referred to this study because its result further supports our assumption that the discrepancy between measured and modelled ozone concentrations at farther downwind source distances is related to chemical processes. We divided the vertical column into a layer close to the ground, two layers in the boundary layer and one layer in the free troposphere. All processes contributing to the mass balance of a species are time dependent and the results are only comparable if the volume considered in the calculation is the same over the whole simulation period. The thickness of each layer had been chosen with respect to the boundary layer structure during the afternoon, where the aircraft measurements took place. Only vertical grid levels of the DRAIS model can be chosen as bottom and top surfaces of a volume. Therefore, a height of 75 m is the best approximation for

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the surface layer during daytime. In the answer to referee #2 further comments to the mass balance calculations are given.

In this context we can answer the question concerning the averaging time used for the determination of the ozone net production rate simulated for greater distances in the ozone plume downwind of Berlin, and which is compared with the corresponding rate derived from the aircraft measurements. The aircraft flew in the lower mixing layer (layer 3 of our mass balance calculations) and it crosses the region 3 only during two short time periods. Therefore, we averaged the simulated ozone net production rates for layer 3 in region 3 over the same periods. This averaging procedure is also explained in the response to referee#2.

The slight modification of the parameterization of the deposition has nearly no effect on the results. It is only necessary to get an adequate “adjoint” deposition. In most models this modification is not necessary. We omit a detailed explanation of the modifications, because it seems to be more confusing than helpful.

Section 3.1.4, describing the tests of the code of the adjoint model, will be modified in order to avoid misunderstandings.

In section 3.2 we introduce a factor FacP instead of the original parameter P by the relation

P = Po* FacP

Po in this equation is now a constant representing the original value applied in the DRAIS model. This transformation is helpful for those parameters, which are time dependent like the emissions. By using FacP instead of P the sensitivity depends only on the level and not on the shape of the diurnal variation of parameter P. For a better comparison of the sensitivities this transformation was applied to all parameters. In the revised version we will add this argumentation.

Section 3.2.1 will be completely revised because sensitivities integrated over the whole

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domain will be added. This has been recommended by referee #2. The integrated sensitivities allow a linear combination of the individual sensitivities. For example, the sensitivity of the distance function related to NOx can be calculated by adding the corresponding values for NO and NO2.

The referee argues about the titration of ozone by NO. His argumentation is correct but we don't see the relevance referring to the sensitivities calculated in this study. The distance function in our study is based on ozone concentrations at the stations Eichstädt and Menz. Both stations are located far from the main sources. And the referee correctly states that the titration is fast and that it plays a role only over the source region.

In cases where it is possible, chemical formulas will be used instead of numbers or other acronyms.

The caption of Figure 2 will be replaced by: "Wind and ozone concentration fields Ĕ ".

The word "chapter" has already been replaced by "section" in the internet version of the paper.

The final remark about the adjoint method is correct. The sensitivity is only a gradient. The absolute value of the sensitivity determines only the relevance of the related parameter. In order to get an acceptable agreement between the measured and simulated ozone concentrations at the stations Eichstädt and Menz, the necessary variation of the parameters has to be determined by data assimilation. In this process the sensitivities are applied stepwise as gradients in order to minimize the distance function.

Discussion of the comments of referee #2:

The referee states that the application of the adjoint model is not generally the most effective method to calculate sensitivities. As he explains, the method depends on the type of sensitivities. But he confirms that the application of an adjoint model is useful for the problem considered in our study. In our understanding, "effectiveness" of the

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method is related to this type of sensitivities and also to the effort of the development of the adjoint model, which by no doubts is a long lasting task. But on the other hand, a comparable time would be necessary if the original DRAIS model would be used to determine all those sensitivities calculated in our study. Moreover, we hope that this model can also be used for further studies.

In the introduction the referee misses a discussion about the adjoint method and its earlier applications. In our paper the adjoint modelling is not the main subject. It is only a tool to calculate the sensitivities. If we follow the suggestions of the referee, we have to go also into more details concerning the data assimilation. In the section about the adjoint model (3.1) the referee recommends further modifications. There, we try to consider the suggestions made in connexion with adjoint modelling.

The second part of the referee's comment on the relevance to other studies (earlier or future) will be considered in the revised introduction.

Referee # 2 also comments on the mass balance calculations (sect. 2.3 of the paper). The reason why we introduced this section has already been explained in our answer to referee #1. For the calculation of the ozone net production rates with the mass balance module of our model system we have selected four vertical layers and three regions along the city plume. We believe that these 12 volumes are sufficient to study the spatial variation of the processes contributing to the mass balance in the plume. As can be seen from Figure 2, the aircraft flew a longer distance in the lower mixing layer above the regions 2 and 3. The chemical ozone net production derived from the aircraft measurements is an average over all time periods the aircraft flew above both regions. Based on the results of the mass budget calculations for layer 3 we estimated a corresponding average for both regions separately. We averaged the calculated ozone net production over the time periods the aircraft flew above the regions 2 and 3, respectively. Based on the comparison between the simulated and "measured" ozone net production, we conclude that the model underestimates the ozone net production at far distances from the city. We do not postulate that the underestimation of the ozone con-

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centration at the station Menz simulated by the model is caused by this effect. We only emphasize that the reduced production rate might be a reason for this underestimation.

Since the ozone production is calculated in the DRAIS model, we have investigated only the sensitivity of the distance function related to the parameters used in this model. Further arguments for this decision can be found in the discussion of the comments of referee#1.

The parameter deposition velocity is not taken into account. Especially, during daytime, where an intensive vertical mixing in the boundary layer takes place, a reduced deposition of ozone does not sufficiently increase the ozone concentration at the station Menz. We performed a test calculation with a deposition velocity reduced by a factor of 3 in the area between Eichstädt and Menz. The resulting effect at the station Menz was an increase of 2.5 ppb only compared to the discrepancy of 15 ppb we are looking for.

Meteorological parameters can also influence the distance function. The reason why we did not consider meteorological parameters has already been explained in detail in our response to Referee # 1.

As far as possible the mathematical notation in section 3 will be adapted to that given by Ide et al. (1997). The symbols for the variables will also be changed, although we have doubts that sub- and superscripts are easier to interpret by the reader.

The section 3.1 is really quite short. In the revised version of our paper we try to realize the suggestions of the referee. However, the text should still be readable for readers being not familiar with adjoint modelling and its mathematical background. Therefore, we do not want to go into details. But we will refer to corresponding papers, where more details can be found.

The sensitivity is a gradient. Therefore, we alter the notation of equation 5 and subsequent equations in order to avoid misunderstandings.

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The reason for introducing FacP is already discussed in the answer to a similar comment of referee#1.

The questions concerning section 3.2.1 are answered as follows: The maximum sensitivities are defined as the referee assumes in his first question. They are integrated over the time between 5 UTC and 16 UTC. The local maximum sensitivities are selected in order to determine the most relevant parameters influencing the distance function. It is also important to know where in the model domain they occur. Variations of the model parameters at these locations provide the greatest reduction of the distance function. These locations are not discussed in our paper. We will add this information in the revised version. The suggestion of the referee to present additional sensitivities integrated over the whole domain in the corresponding figures for the maximum sensitivities will be taken up by us. We accept the argumentation that these sensitivities are a valuable completion to the local maximum sensitivities. In our answer to a comment of referee #1 this point has also been touched.

The time of initialization for the calculation of the sensitivities is 5 UTC. This initialization time was chosen because it is short after sunrise, when daytime chemistry is activated. Additionally, the ozone concentration calculated at the station Menz is still close to the measured value. Deviations from the measurements occur only later during the day.

The referee argues that it would be of interest to start the initialization earlier to reach steady state values. We agree that this may be an interesting aspect. But this additional great expenditure cannot be carried out in the scope of this study, especially with respect to the fact that 5 UTC seems to be a reasonable initialization time. Moreover, it is not sure that new knowledge can be achieved with respect to the aims of the study.

Some of our interpretations of the sensitivities are criticized by the referee. We will try to improve our explanations in the sense recommended by the referee.

The referee comments on errors in photolysis rates. Our study shows that the distance function strongly depends on two photolysis rates only. This is valid also for

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the sensitivities integrated over the whole domain. Therefore, it is easy to perform the comparisons the referee likes to see.

The referee asks for the reason of the variation of the relevant parameters. After having calculated the sensitivities we know the most relevant parameters influencing the distance function. But it cannot be decided, which parameter is most likely responsible for the observed discrepancies in the ozone concentrations at the station Menz. In order to answer this question, we carried out simplified data assimilation. In this context simplified means that a background term in the distance function is not considered.

There are three reasons for using simplified data assimilation:

- It is a much greater expenditure to carry out full data assimilation.

- The error correlation matrix in the background term is not (well) known.

- Because of the missing background term the distribution of the parameters altered by simplified data assimilation may be less smooth than that calculated by complete data assimilation. But the main variations should be similar. After the data assimilation it has to be decided how far the parameter variations are realistic. Basis for this decision are the sign, the location in the model domain, and the amount of the dominant variations. Such a decision should also be possible with the results of the simplified data assimilation.

The results of the simplified data assimilation show that the question, which parameter is mainly responsible for the observed discrepancies, cannot unequivocally be answered. We will discuss this conclusion in more detail in the revised paper taking into account suggestions of the referee. As far as we know, photolysis rates have not been measured during the episode considered in our study.

We do not agree with the comment of the referee on the emissions. If the NOx emissions used in the simulations are overestimated, it cannot be concluded that the same overestimation is also valid for another day. The emissions may vary from one day to

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another as the Augsburg experiment has shown (Panitz et al., 2002). The modelled emissions used in the simulations can be higher or lower than the real ones.

The recommendation of the referee to apply the “corrected” parameters in simulations for other episodes cannot be guided. It cannot be expected that exactly the same parameters have to be corrected in the same manner under other meteorological and background conditions in order to achieve more realistic results of the simulation. We do not believe that there is a general deficit in the parameters of the chemistry model.

Strictly speaking, the results of this study are of course applicable only to the DRAIS model and the specific episode. Although we could not decide clearly which parameters are responsible for the observed discrepancies, the procedure is also applicable to other models and episodes. Discrepancies between measured and simulated species concentrations are often found and they are not a typical behaviour of the KAMM/DRAIS model system. In other cases the results of such a study may give more evidence than the results of this study. The ranking of sensitivities for other chemistry transport models and other questions in context with ozone concentrations may be different, but the relevant parameters are largely the same, because they mainly determine the ozone chemistry in such type of models. Therefore, it is not necessary to repeat the whole sensitivity calculations. They can be restricted to the relevant ones derived in this study.

At the end of the review the referee comments on language and minor issues. These helpful recommendations will be considered in the revised paper. References:

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