Atmos. Chem. Phys. Discuss., 7, S9331–S9344, 2008
www.atmos-chem-phys-discuss.net/7/S9331/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on “Nitrogen Oxide biogenic emissions from soils: impact on NO_x and ozone formation in West Africa during AMMA (African Monsoon MultidisciplinaryAnalysis)” *by* C. Delon et al.

C. Delon et al.

Received and published: 21 February 2008

Response to referees for manuscript acp-2007-0457.

Changes in soil NO emissions. We would like to thank both referees for their useful and constructive comments that helped to improve the quality of the paper, and the reliability of the algorithm. While addressing the referee’s comments, we have found an error in the ANN algorithm. This error was rectified and the agreement between modelling and measurements is now much improved (as illustrated by Fig 7). Furthermore, as both referees stated that the results were "promising but not convincing", we have tried to explain more clearly where we consider there to be good agreement between the model and observations, whilst also being open about the discrepancies. We have clarified that this study is restricted to two days of simulation in the Sahel region. The results are compared to Yienger & Levy approach in the same region, and we try to convince the reader that the ANN approach gives a better result in that particular case. But it is worth noting that we do not (and we cannot at the moment) provide an inventory at the global scale, as provided by YL95. In previous model test runs an amplitude coefficient of the ANN algorithm had been reduced as part of a sensitivity test and it was mistakenly not returned to its correct value. The emissions had then the same range of amplitude as YL95 inventory in the first version of the paper. This coefficient has been put now to its correct value and the algorithm gives much higher fluxes (figure 2). As a consequence, the NOx and ozone concentrations are increased near the surface, and the comparison with measurements is more credible. Accordingly with referee #2 comments, we also have tried to coordinate this paper with Stewart et al. paper from the same issue, by first changing both titles, and by giving inter connected results from both measurements and modelling studies.

Response to referee #1 (S6651-S6654)

Spin-up period: The spin up period is effectively a crucial point. We thank the referee for pointing this out. Concerning the dynamics, a one-day spin-up has been shown to be sufficient for the triggering of mesoscale convective systems as shown by Chaboureau et al., (2007). This sentence has been added in the paper for clarification. The simulation lasts 48h. The analysis of simulation results is made at 15h UTC the second day, 39h after the beginning of the simulation, which can be considered as sufficient for the evolution of chemical compounds given the initialisation method discussed below. The choice of 48h was driven by the necessity of providing the best meteorological and dynamical results. A longer spin up period would have had the inconvenient of degrading the meteorological prediction. A CTM will keep the large scale analysis as its own meteorology, but MesoNH is not a CTM, it is only forced at the boundaries by the large scale forcing every 6 hours, and generates its own meteorology, which can diverge if the spin up is too long. The meteorological prediction was not as good in a 3 or 4 days simulation, the 2 days simulation was then kept. Furthermore, the simulation begins 18h before the convection appears in the domain. Soil surface moisture in the Sahel is very low, and comes back to initial conditions very rapidly after a rain (Taylor and Ellis, 2006; Patricia DeRosnay, personnal communication), in a time scale of 2 days. These comments were added in the MesoNH model description

Chemical initial conditions: The model run was effectively poorly described; we agree with the referee and have changed the manuscript accordingly. The following text has been added in the model description: "The chemistry scheme includes 37 chemical species and 128 chemical equations (Crassier et al. (2000), Tulet et al. (2003), Suhre et al. (2000)). The vertical profiles of ozone, CO, NOx, PAN, isoprene and monoterpenes are initialized from nocturnal average profiles (the simulation begins at 00hUTC) deduced from the idealized 2D modelling study of Saunois et al. (2008). In this study, the MesoNH model was used to recover the typical monsoon regime and the associated distributions of ozone and its precursors over West Africa. Starting from these profiles, the sensitivity runs are shown 39h after the beginning of the simulation (6th August 15:00˜UTC). Methane is initialized to a typical background value of 1700 ppbv. All other species were set to very low values ($<$0.001 ppb). While looking at chemical compounds evolution in the whole simulation domain, this study is focused on chemical processes occurring in the northern part of the domain, the Saunois et al. (2008) initial profiles were therefore averaged between 13 and 15N, and the whole simulation domain receives the same initial values.";

Following the referees comments we have reviewed the initialisation used. Output from a low resolution global model would have introduced an additional source of uncertainty. We have therefore taken initial profiles from a very recent study made specifically over West Africa using a 2-D model. This model has been evaluated through comparison with the observations and been shown to compare well for many compounds. The 2-D model simulation used a 30-day integration spin-up such that it reached a state of

ACPD
7, S9331–S9344, 2008

equilibrium. We therefore consider that, since the chemical initialisation is already the product of a long integration, the 2-day spin-up used for the Meso-NH 3-D model for dynamical reasons (see above) is also adequate for the chemistry. The referees will note however from this new version of the paper that despite realistic chemical initialisation, the ozone in the model is underestimated compared to measured concentrations. This is partly a consequence of the initialisation but also the NOx dilution in the boundary layer has an influence, among other things. As mentioned by the referee, the initial profiles are influencing the chemical state of the troposphere, but in this study we want to show the influence of the NOx emission in a relative way (enhanced ozone concentrations associated with enhanced NOx concentrations over wetted soils, and increased ozone concentrations compared to the simulation using Yienger & Levy (1995) static inventory), not with absolute values.

Canopy Reduction factor (CRF): After checking the description of files provided by the GEIA database, we arrived at the conclusion that the Yienger & Levy inventory is actually provided with an included uptake by the vegetation, as stated by Ganzeveld (http://www.geiacenter.org/) "The inventory by Yienger and Levy [1995], which is available to the modeling community through the GEIA site, is based on an empirical model that accounts for different biomes, pulsing, which is the enhancement of the emissions through rainfall, and the effect of the canopy uptake." This mistake was corrected in the text. It is therefore possible to compare directly with canopy reduced fluxes from the ANN algorithm. The calculation of CRF in the ANN case and of the deposition velocity for chemical compounds (NO2 included) has been described more thoroughly, as asked by the reviewer: "The general resistance parameterisation for dry deposition velocities of Wesely and Hicks (1977) has been introduced into Meso-NH-C (Tulet et al., 2003). The surface resistance incorporates both the physical and biological surface characteristics together with the solubility of deposited species (Baer and Nester, 1992). For vegetated surfaces (Wesely, 1989), one further considers the relative contributions of stomatas, mesophyllic tissues, and cuticle whereas for liquid surfaces, Erisman and Baldocchi (1994) parameterisation is used. These parameterisations have

ACPD
7, S9331–S9344, 2008

Interactive Comment

been included in ISBA and coupled with the 255 surface classification types of Meso-NH. ISBA calculates such evolving parameters as aerodynamical, quasi-laminar, stomatal resistances, and drag coefficients for different vegetation types. So, chemical dry deposition velocities evolves at each time step together with surface wind, turbulent conditions and chemical specificities (Henrys law solubility constant, biological reactivity (Wesely, 1989)). This deposition velocity calculation is of course applied to NO2. The NOx concentration in the above canopy air is deduced from the net canopy emission, minus the above canopy deposition flux. The deposition of NO2 has already been described, and the net canopy emission is a fraction of the upward NO flux from soils. This fraction has been roughly evaluated in MesoNHC, considering that it is a linear function of LAI (Leaf Area Index), derived from empirical relationships between LAI and NO emission (Yienger and Levy (1995) and references therein, Ganzeveld et al. (2002)). The CRF equation is expressed as follows (while using Yienger & Levy (1995) data, it is different from the one developped in their paper), and is valid in a range of LAI from 0 to 8 (m2/m2). CRF= -0.0917 * LAI + 0.9429 This equation does not take into account either the in canopy turbulence, or the leaf resistance to NO2 and O3, mentioned as important parameters in forested areas (Jacob and Bakwin , 1992). Our choice to simplify this equation leads of course to an approximation of emissions to the atmosphere, but the attenuation is almost efficient in rain forests where the canopy is dense, which is not the case in our domain. In our simulation domain, the LAI does not exceed 3.5 m2/ m2 (Fig. 1c), leading to a decrease in NO flux reaching 40% at the most. It is important to note here that this study is not focused on CRF parameterisation, hence our choice for a simple parameterisation. Furthermore, the focus of this study is in the Sahel region (North of 13°N), where the vegetation cover (deduced from the ECOCLIMAP database (Masson et al., 2003)) is below 20%."

p15165, section 5: CTRL run includes anthropogenic emissions. The comparison between CTRL and YL95 runs, and then between CTRL and SOILNOX runs leads to the conclusion that anthropogenic emissions are far lower than biogenic emissions in the region of simulation. Initial ozone fields have been described in the model description in the new version of the paper. Ozone and NOx fields are provided by Saunois et al. (2008), as well as PAN, CO, isoprene and monoterpenes.

The sensitivity calculations are provided by the comparison between the 4 simulations, by adding NOx sources one after the other (anthropogenic, soil and lightning). Biomass burning emissions were not present during the wet season.

Soil moisture and strong NOx emissions: Meixner & Yang (2004) state that molecular diffusion accomplishes the transport of NO in soil pores. Water-filled pores form strong barriers to the emission of NO into the atmosphere. The soil water content also impacts strongly the diffusion of O2 into the soil and consequently the microbial activity (Skopp et al., 1990, added in the reference list). Nitrification and denitrification are intimately related to the soil water content for two important reasons : (a) the substrate supply for soil microorganisms (e.g., NH4+ for nitrifying bacteria) is accomplished by diffusion of the substrates in soil water films, and (b) water in soil pores is the dominant controller of gaseous diffusion in soils (Meixner & Yang (2004)). To make the discussion clearer, the following text has been added in the paper: "The microbial activity in the soil, responsible for NO emissions throughout the soil layer, is influenced by the physical properties of the soil, which affect substrate diffusion and oxygen supply (Skopp et al., 1990). The choice of these seven parameters as inputs has been made to give an insight into these microbial processes, without describing them in detail, but by trying to highlight the different physical processes favouring this microbial activity."

The ANN algorithm does not describe exactly the microbial processes in the soil. ANN uses the examples given in the database to learn how fluxes will have to react when WFPS is high, for specific conditions of pH, temperature, sand percentage, fertilization rate and wind speed (the 6 other inputs of the database). For example, if soil temperature is above 30°C (i.e. tropical conditions), pH is 6 and sand percentage is 50%, the ANN learns to enhance NO flux if WFPS is above a certain threshold (as reproduced by the measurements given as examples in the database).

ACPD
7, S9331–S9344, 2008

Interactive Comment

Comparison with aircraft data and validation of simulated results: As asked by the referee, we have tried in the new version of the paper to be more precise in the comparison between model and measurements. Fig 7 provides NOx low level concentrations for both model (YL95 and ALLNOX simulations) and measurements, in the same location and at nearly the same time (15hUTC in the model, as modelling outputs are available every 3h, and between 14h50 and 15h30 in the observations). Observed NOx concentrations were available only at low level legs, because the sensors did not work correctly on the soundings (concentrations were too close to detection limit). Concerning ozone, the comparison of modelled concentrations with observations shows an underestimate of the model. Ozone low level concentrations are plotted in the paper in fig 9 to compare the respective performances of Yienger & Levy inventory and ANN algorithm for ozone formation. As mentioned at the beginning of the response to referees, the model underestimates the measured ozone concentrations. This is partly due to the initial conditions used in the model. However, the main conclusion from the modelled/observed comparison is that the ANN algorithm reproduces the ozone enhancement consecutive to NOx enhancement over wetted soils (which is not reproduced by YL95), and reaches higher ozone concentrations than the YL95 simulation. The following text has been added: "ALLNOX gives a better representation of the observed ozone concentrations. Both model runs underpredict the concentrations of ozone with ALLNOX giving values of between 30 and 34 ppb and YL95 values of between 24 and 27 ppb whilst the observed are 37 and 44 ppb. However the ALLNOX simulation reproduced more accurately the enhancement of ozone over the wetted patch as seen in the observations"

Language: English native speakers, co-authors of the paper, have corrected the English.

As asked by this referee, the model description has been developed, and the results of the ALLNOX simulation have been compared more thoroughly to observations, with a closer collaboration with Stewart et al. (2008) observed results.

ACPD
7, S9331–S9344, 2008

Concerning the sensitivity runs, each of the 4 simulations considers a different source for NO emission. Anthropogenic sources have a very limited effect on ozone formation compared to biogenic emissions, and are therefore present in all simulations.

Minor comments P15156: abbreviations have been removed from the abstract. L23: ok L25: ok P15159: description of the model has been rewritten. P15160: the sentence has been changed. P15162/3: poorly defined has been changed into "few in situ measurements are available" P15162: ozone measurements are described and are now included as results in figure 9. P15163: The sentence was badly written. pH is not connected to soil moisture. The sentence has been rewritten in:"This moisture effect and the latitudinal distribution of pH values, (lower in the south, as shown in Fig. 1a), reinforces the stronger emissions in the south" L26: sand percentage and strong emissions. This sentence has been added in the text: "High sand percentage results in a higher evaporation rate and so the water content of the soil does not remain high enough to favour the microbial processes responsible for NO emission. Indeed, soil microbial activity is influenced by soil water content and by soil physical properties (regulating aeration-dependant microbial activities important to nutrient cycling, (Skopp et al., 1990))" P15169: First recommendation for further work is to test the ANN algorithm at the annual scale, on a larger domain containing West and Central Africa, to test its sensitivity during the dry season. Second recommendation would be to study more thoroughly the dynamic conditions of the two days studied and to try to understand the underestimation of ozone concentrations. The following text has been added in the conclusion: "The current neural network is however limited due to the sparse observations on which it is trained. More measurements of NO fluxes together with environmental variables (soil characteristics and wind speed) are crucially needed over various regions and seasons. Such a measurement effort should be done in order to provide a large comprehensive dataset linking NO emission to the environment. In particular, a larger dataset would yield a more robust training of the neural network. This algorithm will be used in the future in surface modelling to test its validity during all seasons in West Africa (annual cycle of emissions) at the regional scale. The im-

ACPD

7, S9331–S9344, 2008

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

provement and development of this algorithm could lead to several types of algorithm, depending on the type of climate and soil. In the long term, it would be possible to provide an estimate of biogenic emission at the global scale, and therefore improve the estimate of ozone formation and budget" P15179: ok.

Response to referee #2 (S7775-S7782)

S7776: Hombori experiment: The Hombori experiment (15 June, 15 July 2004) was part of the preparation of the AMMA campaign (Special Observation Period in 2006). During this experiment, NO fluxes were measured at different places around Hombori, and soil samples were collected to analyse the soil texture and pH afterwards. Soil temperature, moisture and wind speed were collected by scientific people from the CESBIO (Centre d Etude Spatiale de la BIOsphere, Toulouse, France) from a meteorological station based in Hombori. But neither ozone nor NOx concentrations were sampled at the surface or in aircraft. This database was used to train the neural network, but cannot be used to validate the ozone concentrations in the atmosphere. However, the mechanism of pulse (increased NO emissions consecutive to a rain event), was learnt by the network from these particular field campaign results. Details about this campaign can be found in Delon et al. (2007).

Specific comments: 1: The sentence was changed in the abstract by: "The neural network algorithm allows an immediate response of fluxes to environmental parameters, on the contrary to fixed emission inventories." 2: ok 3: The condition $[NO]/[O3] > 2.10e\text{-}4$ is satisfied in the case described in the text. The sentence was therefore not modified. 4: Pilegard et al. (2006) was added. 5: Ganzeveld et al. (2002) was added in the text. 6: "The structure and evolution of the boundary layer is determined with an eddy diffusivity turbulent scheme with 1.5 order closure for pronostic turbulent kinetic energy (Cuxart et al. , 2000)."; was added in the model description. 7: the initialisation of the chemical profiles was changed and based on a 2D study in West Africa from Saunois et al., (2008). This was previously described in the response to referee #1, in "chemical initial conditions" p1 and "p15165 section5" p2. 8: ok 9: sentence changed into "To be able to represent different situations 10: In theory, the network can be applied in the range of the data used in the database constituted for the learning, whatever the climatic conditions. Outside this range, it is necessary to be cautious with the results. The purpose of this study (among others) was to actually assess the good behaviour of this algorithm under tropical conditions, and to check effectively if the mixing of temperate and tropical data could be applicable whatever the climate. Finally we (have tried to) prove that the results are convincing, and that realistic NOx levels could be reached by applying this algorithm in the Sahel region. 11. To explain the conceptual meaning of sums and weights, the following sentence has been added in the ANN description (§2.3):"ANN are built by analogy with the human brain. The learning of the human brain is vital for its development, and contacts between neurons (i.e. transmission of the information) are provided by the synapses. In ANN, the synapses are represented by the weights affected to each input parameter. The link between inputs (with their affected weights) is made through a mathematical (activation) function." The seven parameters used in the equation are described as follows in the ANN description (§2.3): "The microbial activity in the soil, responsible for NO emissions throughout the soil layer, is influenced by soil physical properties, which affect substrate diffusion and oxygen supply (Skopp et al. , 1999), The choice of these seven parameters as inputs has been made to give an insight of these microbial processes, without describing them in detail, but by trying to highlight the different physical processes favouring this microbial activity. The network was initially run with soil surface temperature and WFPS as inputs, because of their well known and fundamental influence on NO emissions, reported in all of the literature on the subject, cited in the introduction of this paper. Soil temperature at depth was includeddue to the effect it has on oxygen diffusion and N mineralization into the soil (Butterbach-Bahl et al., 2004). Fertilisation rate gives the amount of nitrogen input (natural and/or anthropogenic), in part responsible for the rate of gaseous emission at the surface (Sanhueza et al. , 1990). Sand percentage is an important feature for emissions through its link with water diffusion (Roelle at al. , 2001). pH conditions can influence NO emissions via chemical or biological processes

ACPD
7, S9331–S9344, 2008

(Serça et al., 1994), Wind speed is a way to represent the state of the atmosphere at a given time (Delon et al., 2007)" 12. To improve the clearness of the CRF parameterisation used, the section on CRF has been updated. In the first version of the paper, the description of YL95 emissions was misunderstood by the authors. Actually, the YL95 inventory had a canopy uptake included, and could be directly compared to the algorithm with CRF. To understand the details of this parameterisation, and the text that has been added in this new version of the paper, the referee #2 is kindly asked to refer to the response to referee #1, chapter "Canopy Reduction Factor", pages 1 and 2 for more details. In our opinion, the upscaling of ANN simulations to 1°/1° and one month is not justified in that study, because the main advantage of the ANN algorithm is its ability to give an immediate response to rain events, by being coupled on line in the chemistry-dynamics MesoNHC model. If the emission is averaged throughout one month and one degree, the resulting flux is smoothed (see comparison between YL95 and ALLNOX signals on ozone and NOx concentrations in Figures 7 and 9). However, the spatial upscaling would be justified by the use of a CTM at a broader scale, but keeping a high time resolution (1 day to 10 days) would be necessary to reproduce the rain impact on fluxes. 13: Indeed, the drying of soil surface in the Sahel is very fast. The following sentence has been added in the text "Indeed, the drying of soils in the Sahel is around 1-2 days (Taylor and Ellis, 2006). NO emissions from sandy soils have been found to decrease rapidly, over 2- 3 days after wetting (Scholes et al., 1997; Johansson et al., 1988)". The following response has been given to referee #1: "Soil surface moisture in the Sahel is very low, and comes back to initial conditions very rapidly after a rain (Taylor and Ellis, 2006; Patricia DeRosnay, personnal communication), in a time scale of 2 days." 14: As suggested by the referee, the results of Ganzeveld et al. (2002) for the flux in the Sahel region have been added in the text. The previous calculation made over West Africa was removed and restricted to the Sahel region, in accordance to Stewart et al. (2008) paper. 15: The definition of CRF and NO2 deposition onto plants has been developed in the model description. 16: To be as clear as possible, the equation has been added in the new version of the paper. This specific question

ACPD

7, S9331–S9344, 2008

Interactive Comment

about CRF has been detailed in the response to referee #1, and will be hopefully sufficient to answer referee #2. 17: Suggested change in vocabulary has been made. 18: A map of LAI has been added in figure 1c, giving an information of the spectrum of vegetation in the simulated domain. The CRF is not a diagnostic variable in the model, and CRF map is not provided as a 2D parameter. 19: To improve the understanding of the text, the sentence has been changed by "Fig. 3a, (YL95-CTRL) shows the impact of biogenic emissions from YL95 inventory, compared to anthropogenic emission only" 20. This sentence has been changed in "Figure 3b shows the concentration difference between SOILNOX and CTRL simulations, showing that the introduction of ANN on line emissions coupled with CRF increases the NOx concentrations in the lower troposphere (up to 700 ppt) stronger than the anthropogenic emissions of the CTRL simulation do." 21. As mentioned in point 3, the condition [NO]/[O3] $>$ 2.10e-4 is satisfied in the case described in the text. The sentence was therefore not modified. 22. ALLNOX-CTRL and SOILNOX-CTRL will give nearly the same result between 0 and 2000m, because the impact of NOx from lightning in ALLNOX is only visible between 10 and 15 km height. Before commenting each difference, a short sentence has been added to clarify what we wanted to highlight. 23. "verified" has been changed into "shown". 24. Indeed, the sentence mentioned here was the result of a discussion with people working on LiNOx. This sentence has been changed, as we could not provide any reference for it. It is now "Indeed, from three years of satellite measurements, it is apparent that while all continental regions have an afternoon peak (1300-1500 LT) in deep convection for non-MCS events, Africa's MCS convective intensity rises to a maximum level between 1900 and 0500 LT (Nesbitt and Zipser , 2003). Here the simulated convective event is the most intense at 2100 UTC (NOx differences range from 0 to 2 ppb around 9-14km altitude, whereas the maximum O3 difference is situated around 6-8km, and reaches up to 9 ppb." 25. subheading 6.1 was removed 26. This sentence was corrected 27. 500m above sea level (ASL) 28. South west and north east became south and north respectively. 29. Figure 8 was kept to show the latitudinal cross section from south to north with altitude. In this new version of the paper,

ACPD

7, S9331–S9344, 2008

Interactive Comment

figure 7 shows the comparison between observed and modelled NOx concentrations in YL95 and ALLNOX simulations, and Figure 9 shows the same comparison but for ozone. The emissions have increased by correcting one coefficient in the algorithm. A factor 1.5 remains between the modelled and observed NOx increase above wetted soil, but it becomes 3.4 when the YL95 inventory is used, and the concentrations do not increase with wetted soils. The enhancement of NOx is reproduced by the ANN algorithm and concentrations are approaching observed levels. In our opinion this is an important improvement over the YL95 static emission inventory in that particular case. 30. The range of measured concentrations is now better reproduced by the model, as shown by figure 8. The 20 km resolution remains important to keep the enhancement of NOx concentrations over wetted soil. A spatial resolution of 1° would produce inferior results. Of course we are not reproducing the measured values exactly, but the purpose of this paper was first to show that the ANN algorithm was able to reproduce NOx concentrations enhancement when the rain falls in Sahelian climate, and that the background concentrations were also in the range of realistic concentrations. For that, several simulations have been performed to show the difference of impact of different sources of NOx. To validate these results, and to show that the algorithm is reliable, we have used observed data. Now, several improvements can be (and will be) brought to this algorithm and its application would become even more accurate. 31. The authors think that it is worth discussing that the slight increase from south to north is not reproduced by the model, even if a factor 2 (or less) is remaining between observations and model results. This is the beginning of new ideas and corrections that can be applied to the algorithm, to improve its impact, onto vegetated cover for example. 32. The top of the boundary layer is 850m in the observations, and is 1000m in the model. This was added in the text. 33. The boundary layer heights are not too different (850m in the observations vs 1000m in the model). The big difference between the model and measurements lies in the stratification between the boundary layer and the layer above. In the observations, ozone is well mixed from 0 to 4000m. In the model, ozone concentrations increase in the boundary layer. Furthermore, as written in the text, the

ACPD

7, S9331–S9344, 2008

turbulent scheme in the model above wetted surface may react too weakly, and may lead to a slow and limited diffusion of chemical species from the surface to the top of the boundary layer. For these reasons, the NOx and ozone concentrations are lower in the model than in the observations. 34. Accordingly to referee comment, Figure 11 was corrected. 35. The ANN algorithm is an improvement on YL95 because it gives NOx enhancement above wet surface. For the same initial conditions, same domain and same days of simulation, the YL95 inventory leads to underestimated concentrations of NOx and ozone in the boundary layer. The ANN algorithm does not correct everything, and does not answer all questions, but in this particular region of the world, where the pulse effect contributes strongly to the global budget of NOx, it gives a better estimate than YL95 inventory. On the other hand, YL95 inventory is better everywhere in the world, because the ANN algorithm is for the moment only tested in West Africa. 35bis. This sentence did not have a real sense and was removed 36. In this revised version, the model runs have been changed with new initialisations and the ANN has been corrected (see above). This has led to increased emissions and concentrations in the lower troposphere for the ANN model run. We have also followed the suggestions and taken account of the comments of the referees to improve the presentation of this study. We also have read and referenced the manuscripts suggested by the referee. We believe now that the modelling results show very good agreement with the observations and that we have presented this in a more convincing way.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 15155, 2007.

ACPD

7, S9331–S9344, 2008

Interactive Comment