Atmos. Chem. Phys. Discuss., 5, S5108–S5117, 2005
www.atmos-chem-phys.org/acpd/5/S5108/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "European surface ozone in the extreme summer 2003" *by* S. Solberg et al.

S. Solberg et al.

Received and published: 17 January 2006

Referee #1

The general comments:

Meteorological conditions The referee asks for a more extensive and quantitative meteorological section, such as surface and 850mb pressure, surface temperature, and/or horizontal wind field and how they relate to the surface ozone distribution etc. The referee also suggests to include ozone sonde data.

Response: We agree that the meteorological description was rather short. To avoid expanding the manuscript too much, we have now included a section looking more detailed into the meteorological development during the first half of August when the most intense heat wave occurred and when the Iberian fires took place. We have discussed the day-to-day development in ozone in light of the development in synoptic situation,

winds and temperature on the 850 hPa and ozone soundings from Uccle, Belgium. This required two more figures to be included. Another co-author is also included in the manuscript. Although not very detailed we think this analysis has shed new light on our results and has indicated more clearly the likely reasons for the development of the extreme ozone episode experienced during this period. Data from the vertical soundings have been particularly useful for the interpretation. One could always ask for more stations and longer time periods. We think the present text is an appropriate compromise between detail and manuscript length.

We have also moved the section on residence time in the boundary layer to the section discussing the general meteorological conditions in the beginning of the manuscript, as suggested by the referee.

The specific comments:

1. The referee disagrees with the statement that we have shown a relationship between anticyclonic conditions and residence time in the boundary layer.

Response: We have rephrased the sentence in the abstract, stating simply that we calculated the residence times and found them to be prolonged for some sites. For the comment on meteorology we refer to our response to the general comment above.

2. The referee argues that we need support for the hypothesis that drought is the most contribution to elevated ozone or leave out the hypothesis.

Response: We agree and have moderated our statement. We now include as a hypothesis that the drought lead to reduced ozone loss, but we have taken out the statement that this is the most important process.

3. The referee feels that the statement about PM in the abstract should be removed.

Response: We have removed this statement.

Meteorological conditions See our response to the general comment above

ACPD

5, S5108–S5117, 2005

Surface ozone 1. The referee asks for a comparison of the EMEP data with other data sources. The referee also asks whether the EMEP network has been subject to an evaluation and wonders about the link to national networks.

Response: We have included a bit more text on the criteria for selection of EMEP sites and a number of additional references to the EMEP ozone data being used in the scientific literature. We are, however, not sure how the referee imagines a comparison with other data or a validation. How can one validate data from a monitoring network apart from assuring the best QA procedures? And how specifically should one compare the continuous monitoring data from 100 European sites to a few vertical soundings highly scattered in time and space in a meaningful way? As we don't see any clear answer to these questions, and as we believe that the EMEP ozone monitoring network through its 20 years of history is sufficiently well established, we think our response to this point is sufficient.

2. The referee questions the use of annual maximum hourly ozone values ("an hourly peak throughout the entire year seems to loose its meaning").

Response: The reason for using such an extreme value is to select a parameter which is indicative of the photochemical ozone episodes that year. It is well known that such episodes are most clearly reflected by the extreme values. The annual average ozone values would for example be of little use as this parameter is to a large extent controlled by other processes like background etc. However, we agree that the annual peak value may seem as a radical choice, so to meet the critic we have now used the 99-percentile (based on the daily maximum values) through the year.

3. The referee asks for a reference to the long-range transport event mentioned.

Response: We have reformulated this and now just mentions the ozone concentrations observed. (The episode mentioned is part of a manuscript in preparation).

4. The referee requests the purpose of the discussion of the relationship between low

ACPD

5, S5108–S5117, 2005

humidity and ozone generation.

Response: We have tried to clarify this by reformulating the text in the manuscript. The purpose is to show that some feedback mechanisms between high-pressure systems and ozone formation may actually be negative.

5. The referee finds the blue lines difficult to distinguish.

Response: We agree, and have changed them in the revised figures

Residence time The referee suggests to link this section with the preceding section on meteorology.

Response: That is now done.

CO/Siberian fires The referee suggests that we combine the section on CO/Siberian fires with that of the Iberian forest fires. The referee questions the benefit of Figure 9. The referee thinks we should remove the sentence about the link between global warming and tropospheric ozone.

Response: We think it is a good advise to combine the two sections and have done so in the revised manuscript. We also agree that the signal of elevated CO in the summer 2003 measured at Spitsbergen is modest compared to the other years and not convincing by itself. However, we believe the measurements from Weathership M do show a fairly marked increase in CO in summer 2003 (and 1998) compared to the other years which is relevant for the discussion and deserves to be published. To let out the Zeppelin time series is not an alternative as that would mean a subjective screening of data. Furthermore, we have reformulated the discussion about a possible increase in the background CO. More references are included that show that it is likely that massive boreal fires may increase the general background level of CO and ozone. Supported by these studies and by the CO data from Weathership M, we conclude that is also likely that such an increase did happen in 2003.

We have also made a link between the discussion between the Iberian fires and the

ACPD

5, S5108–S5117, 2005

discussion of meteorological development during August which indicated transport of warm and ozone rich air masses above the stable PBL during the last part of the August ozone episode.

We do, however, not agree that the sentence about global warming and tropospheric ozone should be removed. We think this is an important point, although – we admit – not proven. Meteorologically, 2003 has been referred to as a signal of what to come if predictions of future climate is correct. The link between the Iberian fires and the drought in 2003 is quite clear, and if one then believes that these fires in turn will produce ozone, an important feedback mechanism of the climate system is established.

Total ozone The referee suggests either to expand the analysis of total ozone quantitatively or take it out. The referee is not convinced that total ozone was particularly low over central Europe or that a reduced ozone column in the north would have any influence on surface ozone.

Response: Although, as stated above, the purpose of this manuscript is to present various likely feedback mechanisms between the 2003 heat wave and increased surface ozone, thus not analysing in detail each individual process, we have decided to take out the section on total ozone.

Forest fires on the Iberian Peninsula The referee suggests to look also at the flow regime from August 4th on.

Response: We think this is now taken care of by the new Figure 9, showing daily maps of surface ozone plus winds and temperature on the 850 hPa. A link to the meteorological section is also made in the discussion of the Iberian fires.

Conclusions The referee has a number of wording issues.

Response: In the "Conclusions" now called "Summary of results" we have rephrased the various statements accordingly.

ACPD

5, S5108–S5117, 2005

Response to technical corrections made by referee #1: All points have been corrected except for the last point (#9): September 2002 is correct.

Referee #2

General comment The referee suggests to change the title from Conclusions to Discussion of results.

Response: We have changed it to "Summary of results"

1. Residence time The referee comments that the residence time is only one of several meteorological conditions required for effective ozone formation.

Response: We agree to this and think that is partly answered through the addition of a section on the meteorological situation during August 2003. We have also included a sentence in the manuscript stating that increased residence time is only one of several necessary conditions for affective ozone formation.

2. Importance of NOx:VOC ratio The referee wonders if the European NOx and VOC emission reductions could actually have lead to a more efficient ozone formation environment by altering the NOx:VOC ratio in Europe.

Response: It is well known that the ozone formation is not only controlled by the absolute level of NOx and VOC but also on their concentration ratio. However, we don't think this could explain an ozone increase in Europe in general. The mentioned paper by Vukovich & Scarborough seems to be based on empirical studies only, not on CTM calculations. The question about the role of NOx:VOC ratios for ozone formation has been extensively studied in EMEP by use of the EMEP model, and the calculations clearly show that a general reduction in ozone is to be expected in Europe from the significant emission reductions that has taken place. In a few smaller regions the emission reductions is predicted to lead to increased ozone levels, but that is mostly confined to the areas close to the strongest emission areas (e.g. the Netherlands/southeast England). This is shown in several modelling reports from EMEP-MSC/W (available online from

ACPD

5, S5108–S5117, 2005

www.emep.int).

3 + 4. Isoprene The referee comments that as we show only one site with isoprene measurements we should be careful with making statements for the whole EMEP region. The referee also thinks we should moderate the conclusive statement on page 9016.

Response: That's a valid point. Actually there are isoprene measurements from a few more EMEP sites, but we chose not to use them in the original manuscript as we believe Donon is the site which is the most influenced by biogenic isoprene emissions. This is quite clear from the general isoprene level itself and also from the knowledge of the surrounding environment. Thus, other sites, further away from main biogenic emission areas, will to a larger extent only show what's left of the biogenics after considerable oxidation of the primary species. This will be true for any species, but for isoprene it's more crucial as the lifetime is so short. Anyway, we agree with the referee that we need to be very cautious with conclusive statements. Not only did the conditions for biogenic emission change in 2003, so did presumably the rate of vertical exchange (higher stability) which would also show up as increased levels of surface emission components. We have included more text in the manuscript mentioning the points above and mentioning that results from other sites exist and are now less conclusive than in the first version of the manuscript.

The conclusive statement about the influence of isoprene on page 9016 has been removed.

5. Total ozone The referee thinks we need more explanation why reduced total O3 should affect PBL ozone.

Response: This section is now taken out of the manuscript.

7. Importance of background ozone The referee thinks that the role of possibly elevated background ozone through extensive forest fires for increasing the formation of ozone

ACPD

5, S5108–S5117, 2005

in the European PBL should be considered in-depth by the authors.

Response: This touches the general concept of the whole paper. The idea of our paper is to bring in a number of topics all linked to the extraordinary conditions in 2003 and show by measurements and transport calculations that many of these topics are likely to have favoured ozone formation. Each of these topics would require a detailed and more quantitative study by its own to give more conclusive and quantitative answers, in other words separate papers for each of them. We agree that it would be interesting to go "in-depth" as asked by the referee on this topic, as well as the other topics. That is, however, beyond the scope and the size of the present manuscript.

8. Iberian fires The referee asks if we have any CO or ozone data to show that the background levels were enhanced when the air mass was from the Iberian fires reached the EMEP region?

Response: A simple answer is no. As shown in the manuscript the surface ozone network is limited, and only a few background sites in Europe monitors CO. Thus, it will be very difficult to distinguish in a convincing way between background (i.e. European inflow of air masses from the Iberian fires) and air exposed to other kinds of emissions during these events. The discussion now included on the meteorology and ozone distribution during August touches this topic to some extent.

9. See "General comment" above

10. Drought/biogenic emissions The referee comments that we can't argue with increased biogenic emissions and reduced ozone uptake due to closing of the stomata due to dry conditions at the same time.

Response: This is a relevant comment. To answer this question unfortunately requires a network of highly specialized field data as soil moisture, biogenic emissions, local flux measurements etc which don't exist except from certain research field studies or perhaps at a few locations in Europe. We have no reason to doubt the measurements

ACPD

5, S5108–S5117, 2005

of isoprene at Donon. Previous studies and comparisons with model calculations have given very good confidence in these data. The most likely explanation as we see it, is regional differences in Europe. Drought in many areas of Europe has been mentioned by papers referred to in our manuscript. Elevated isoprene, either due to high emissions or increased atmospheric stability is also seen, but only at one site, Donon in southeast France, located in a forested area, possibly less sensitive to dry conditions than less vegetated areas.

Our hypothesis, and it is only that, is that depending on the local surface cover, temperature and stability the drought lead to closure of the plants stomata, thereby reducing their ability to take up ozone and emit isoprene in many regions, while in other regions, still above the soil moisture threshold, increased biogenic emissions and ozone uptake occurred. This can only be speculations.

In our revised manuscript we have moderated the conclusive statements about isoprene as explained above. We have also changed our previous statement about the importance of surface drought and ozone deposition. Finally, in the last section we have included a discussion in line with the discussion given here.

Comment by S. Sillman

S. Sillman comments that there is a relationship between high temperatures and efficiency of ozone formation (through the thermal dependency of PAN stability) that may have been an important mechanism in the 2003 heat waves, and suggests that this should be included in the list of contributing factors in the manuscript. He refers to calculations indicating that above approximately 300K, the increase in thermal decomposition of PAN means that less reactive NOx is stored as PAN and more is available for ozone formation, which is of importance in rural areas where ozone is NOx limited. He says that daily maximum ozone increased by 3 ppb for every 1-degree rise in daily maximum temperature (Sillman and Samson, 1995; Wunderli and Gehrig, 1991- these refs are in his comments).

ACPD

5, S5108–S5117, 2005

Response: We are thankful for this comment and think it is a valid point. We have included a statement regarding this in Chapter 3 "Surface ozone" and have referred to the 1995 paper by Sillman and Samson. A sentence about this is also included in the Abstract and in the Summary. This also add on to the list of proposed contributing mechanisms that we unfortunately can't investigate in a quantitative way as that would require detailed photochemical model calculations. As stated above, each of these individual processes would require a separate study including detailed modelling exercises to be more quantitative, which, however, which would exceed the reasonable limit of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 9003, 2005.