Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-1097-RC2, 2021

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Spatial and temporal changes of the ozone sensitivity in China based on satellite and ground-based observations" *by* Wannan Wang et al.

Anonymous Referee #2

Received and published: 18 January 2021

Anonymous Referee #2: Comments to: Wannan Wang et al. "Spacial and temporal changes of the ozone sensitivity in China based on satellite and ground-based observations" MS No.: acp-2020-1097 MS type: Research article Special Issue: Regional assessment of air pollution and climate change over East and Southeast Asia: results from MICS-Asia Phase III

General comments:

The paper provides a substantial contribution to the understanding of photochemical ozone production in China. Instead of looking directly at ozone precursors (NOx and VOCs) in the field, it is demonstrated here that information on the photochemi-

cal regime can be derived from satellite data of NO2 and HCHO column densities. In this context, the authors develop a method to discriminate between VOC and NOX dependence using characteristic monthly HCHO/NO2 column density ratios from satellite observations. An important aspect of this approach is that they contrast satellite-based HCHO/NO2 ratios for a large number of ground-based ozone measurements and derive characteristic thresholds for VOC or NOx dependence of ozone formation. In the last part of the paper, the developed methodology is applied to the exceptional situation of the COVID-19 lockdown in late winter 2020 in China. The increase in ozone concentrations observed for numerous areas in eastern China was contrasted with the parallel observed decrease in NO2 column densities, and resulting changes for the pre/post lockdown photochemical regimes prevailing in China were derived from the satellite-based HCHO/NO2 ratios. The authors conclude by pointing out that China's future ozone reduction strategy should in any case be accompanied by a parallel VOC reduction control in addition to the focus on NOx reductions that has prevailed so far.

The paper is well written and structured and clearly identifies the scientific sources used. The authors make a clear distinction between their own contributions and adequately acknowledge previous work from the literature.

Overall, the following ratings are given: Scientific significance: Excellent (4) Scientific quality : Good (3) (see comments) Presentation quality : Excellent (4)

Special comments:

Line 113: The authors point out that only data from the afternoon are used to determine the HCHO/NO2 ratios in order to describe the peak of photochemical ozone production. However, HCHO is a trace gas that is both emitted and photochemically formed in parallel with ozone production. Would the authors please comment on why they do not distinguish between directly emitted HCHO and photochemically formed HCHO. Shouldn't only photochemically formed HCHO be a measure of the intensity of ozone formation? And would it not be possible to draw conclusions about the proportion of directly emitted HCHO (e.g. from vehicle exhaust gases) from additional "satellite winter data" to be evaluated?

Line 120: The authors point out that they use solar zenith angles of < 80° to determine the monthly HCHO/NO2 column density ratios. However, due to the finite pixel size, the intensity of HCHO production depends on the integral solar radiation lasting for several hours. Therefore, should not different HCHO/NO2 ratios be used due to the enormous extent of the study area (< 20°N - > 45°N) ?

Line 129: The authors point out that the monthly mean values of the HCHO column densities of (0.05° * 0.05°) have been converted to the pixel size for NO2 of (0.125° *0.125°). Can it be excluded that this procedure leads to significant changes in the HCHO/NO2 ratios (after all, the ratio of the column densities of HCHO/NO2 contains the quotient of the precursor (NO2) and the product of photochemical processing. The adaptation of the HCHO pixel size to that of NO2 could lead to a systematic underestimation of the HCHO column densities. Would the authors please comment on this.

Line 175: Using the CLASS model, the authors demonstrate that photochemical ozone production can be represented in terms of O3 isopleths as a function of HCHO (as a proxi for VOC) and NO2. Shouldn't the isopleths rather represent the ozone production over a period of time instead of the total ozone concentrations shown? Elsewhere, the authors explicitly point out that they distinguish between background ozone and additional ozone production by local photochemical ozone production in the measured ozone monthly means (c. f. line 207). It is suggested to describe the non-linearity of ozone production either only schematically (in the form of a cartoon) or actually quantitatively by naming all boundary conditions (starting concentrations, radiation conditions, background ozone concentration, ...).

Line 220: The authors plot the measured monthly means (noon) as a function of the FNR ratio for a large number of monitoring stations. From the summer O3 monthly means > 160 μg/m3 they calculate a median for the FNR (3.28). The 20% and 80%

percentiles are then used as thresholds for VOC and NOx limitation. Would the authors please comment on why it is justified to assume that thresholds can be inferred unambiguously in this way ? For this, the ozone monthly means of the measuring stations used must satisfy a given frequency distribution of the photochemical regimes. (After all, it is conceivable that the Chinese O3 monitoring network contains practically only stations with NOx limitation. Then this approach (i. e. by calculating the 50% percentile of the O3 measuring stations for which O3 monthly means > 160 μg/m3 are observed) would shift the center of the transition range far into the range of NOx limitation). Would it not be more appropriate to argue that the highest photochemical ozone production must occur in the transition region? It is therefore proposed to use an isopleth plot of summertime O3 levels above 160 μg/m3 as a function of HCHO and NO2 column densities to identify the HCHO/NO2 ratio of maximum ozone production. In this way, the HCHO/NO2 ratios for NOx and VOC limitation can be determined independently of the frequency distribution of ozone monitoring stations with values above 160 μg/m3.

Line 315: Are the same HCHO/NO2 thresholds used for the COVID-19 lockdown periods (Jan. 2010, period I and Feb. 2020 period II) from Fig. 6c and 6d as for summer conditions ? It is evident that due to the different radiation conditions alone, the ratio of directly emitted HCHO and photochemically formed HCHO (see also comment to Line 113) is significantly different in late winter than under summer conditions, so that a change in the threshold ratios for VOC- and NOx-limitation should also be expected. It is suggested that the VOC- and NOx-limitation thresholds for the COVID-19 lockdown periods should be determined independently (e.g. using the isopleth method proposed above).

Technical corrections:

Only two minor issues were found when reviewing the manuscript: - Line 68: Please replace "... to the summed rate of reactions of VOC with peroxy radicals". "... to the summed rate of reactions of VOC with OH radicals".

Line 68: The reference (Sillman, 1995) does not appear in the bibliography.

Overall, I consider the approach of the paper a promising way to describe different photochemical regimes. It will certainly be the task of further refinements of this approach in subsequent papers to make even more differentiated statements on the optimisation of ozone reduction strategies. However, satellite-based analysis of photochemical regimes does not seem to be a complete alternative to OBM studies, especially since in the first case the composition of the VOC mix (e.g. the processing of CO and CH3OH has very different HCHO production efficiencies when using the same OH reactivities for both species) is left out.

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