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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Photochemical age of air pollutants and oxidation products in transboundary air observed on Fukue Island, Nagasaki, Japan" *by* S. Irei et al.

Anonymous Referee #3

Received and published: 13 February 2016

Overview: This manuscript presents measurements of mixing ratios of selected trace gases and composition of sub-micron secondary organic aerosol at a rural location in Southern Japan. The authors compare these measurements to previous measurements from this site and other sites in the region, discuss correlations between measured species, and estimate the photochemical age of air masses using concentration ratios of selected trace gases. The stated purpose was to use these age estimates to explain variation in the concentration of secondary pollutants, specifically SOA and ozone. Understanding the evolution of SOA and factors influencing ozone formation are highly relevant areas of research especially in the southeast Asia region. The major strength of this manuscript is the high quality dataset generated at a site that receives

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Discussion paper

air masses from industrialized areas of China after a short transit across the East China Sea. The experimental approach is straightforward, but sound, and the methods used are adequately described. The paper is, for the most part, well written with clear organization. Ultimately, the authors present an independent, quantitative estimate of the relationship between photochemical age and oxidized organic content of SOA; however, the manuscript does not clearly address the stated objective of determining the relationship between photochemical age and ozone mixing ratios. While this relationship is discussed in lines 362-368 and 385-392 and in Figure 10, the manuscript could benefit from a clearly identified and consolidated discussion of this relationship similar to that in section 3.6 for f44 vs. t[OH]. Alternatively, the mention of ozone in line 79 should be eliminated. In addition, several other issues should be addressed to improve the clarity of the manuscript as detailed below.

Specific comments:

Title: The term “oxidation products” could be more specific as many readers would consider this to imply that gas phase oxidation products (OVOCs) were measured. Perhaps mention SOA or use the term “secondary air pollutants”.

Abstract: It may be beneficial to mention the range of t[OH] calculated using NOx/NOy. Given the stated purpose of the study, some mention of the relationship between ozone and NOx/NOy should be included. Can the importance of the calculated f44 increase rate be put in better context in final sentence instead of a simple comparison to the NEAQS data?

Introduction: The sentence on lines 50-51 indicates that air masses move from east to west; this is opposite of the direction from China to Japan. Otherwise, this section is clear and concise.

Experimental: This section is also sufficiently thorough, but also concise. A more detailed description of potential local sources of trace gases in the study area would be beneficial for readers unfamiliar with Fukue Island. For example, are there agricul-

tural operations that could contribute to the high particulate nitrate concentrations, or combustion sources other than automotive traffic that could contribute to ethyne and CO?

Results and discussion: Section 3.1: Lines 139-140: How were precipitation events determined to have a negligible effect on trace gas and aerosol data?

Section 3.2: Lines 167-173: Are there agricultural operations in this region that could contribute to the high particulate nitrate concentrations? Lines 224-228: The seven high-concentration episodes were derived from industrialized areas of the Asian continent. This text should be added because the clean air masses originating in Mongolia are transported from the Asian continent, too.

Section 3.3 Lines 242-243: Ethyne is a tracer for combustion sources in general, not just vehicular emissions. Line 250: Explain what variables were used in the regression. Was it CO and NOy? Lines 253-254: What do you mean by recently improved emission of NOy? Reduced emissions?

Section 3.4 Lines 289-290: What was the match percentage for your HOA and LV-OOA spectra compared to the Ulbrich database? Lines 299-300: What does the high OM/OC ratios similar to humic-like substances say about the sources of your observed OA?

Section 3.5: Temperatures in this section are given in K and °C. Choose one unit and be consistent throughout the manuscript. Also, at some point in this section, it is important to explicitly state that no reliable t[OH] was calculated from the hydrocarbon clock. Lines 313-315: Consider rephrasing these sentences to indicate that because there are few potential sources of these gases between emission sources in Asia and the study site, this study offers an opportunity to use photochemical clock estimates under nearly ideal circumstances. Lines 374-376: The difference is consistent with what? Please clarify your meaning. Lines 446-447: These values refer to “natural log-transformed hydrocarbon ratios” Line 498: Significantly low what? It looks like

something is missing in this sentence.

Section 3.6: This discussion could benefit from a clear statement comparing the proportions of HOA and LV-OOA observed in the 2014 measurements (Irei et al. 2015) and in the current study and the correlation coefficient with t[OH] or NOx/NOy for each dataset. This may be helpful in determining a minimum proportion of LV-OOA necessary to use f44 as an indicator of oxidation.

Summary: Again, the use of the term "oxidation products" could be more specifically referred to as oxidized organic particulate matter, and some mention of the relationship between ozone and t[OH] should be made.

Figures: Figures 2 and 3: Can these be combined to include wind direction in Figure 1? If not, it would be helpful to have percentages on the wind rose in Figure 3 to indicate the distribution of wind direction observations. Figure 5: This figure is quite large. Can panels with species with similar concentration ranges be combined? Also in panel (e), why does the baseline concentration of isopentane decrease after the break in the data? Was this a calibration issue? Figure 6: A boxplot of CO mixing ratios binned by wind direction may be more useful in demonstrating the lack of wind dependence. Or adding a mean or median line to the wind rose would help. Figure 12: It would be helpful to show an overall trendline for the data to allow for a comparison with the modeled trends.

Tables: Double check that consistent significant figures are used in all tables. In Table 1, for example, the Max CO mixing ratio is given to 4 significant figures, the median to 3 sig figs and the minimum to 2 sig figs.

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