Atmos. Chem. Phys. Discuss., 12, C7391–C7397, 2012
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Atmospheric Chemistry and Physics Discussions

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12, C7391–C7397, 2012

Interactive Comment

Interactive comment on "Contrasting trends of mass and optical properties of aerosols over the Northern Hemisphere from 1992 to 2011" *by* K. Wang et al.

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Received and published: 25 September 2012

Introductory Remarks Comment: The authors use regional databases for the Northern Hemisphere to compare PM10 and PM2.5 mass concentration data with daily mean visibility observations which are then converted to optical extinction coefficient values. The work is carried out for 4 regions over the Northern Hemisphere– that of Canada, China, Europe and the US. The result of a decreasing PM10 over the past decade or more is already fairly well established from sources such as National and Regional Environmental Agencies. The findings regarding PM2.5 variations since the mid to late 1990's are more interesting, as is the decrease in visibility for 3 of the 4 regions examined.

Response: Thanks. Below is our point-to-point response to your comment.

General Comments 1) Comment: Since PM2.5 is a measure of the mass of aerosol particles of size which are the dominant influence on optical extinction coefficient, a serious omission from the paper is an investigation of annual anomalies for PM2.5 – which is presented for PM10 in Figures 9 and 10. This might have provided evidence in support of the statement made in the paper [Page 17923, Line 16-21], which infers an increase in number concentration of fine smoke particles (and a consequential increase in PM2.5 mass concentration) due to increased number of wild fires in the US. However having said that, while Figure 6 shows fairly constant PM2.5 levels over the period from about 2002 to 2007 (with the exception of 2005), an overall decrease in PM2.5 levels occurs over the period from 1998 to 2010. In summary, the explanation given is not entirely convincing.

Response: We added and discussed PM2.5 to Figures 9 and 10 in the revised paper. We deleted the discussion on the impact of fires in the U.S.

2) Comment: It is also not clear why monthly anomaly plots analogous to the Figure 9 and 10 annual anomaly type plots were not presented or discussed.

Response: When using Figs. 9 and 10, we tried to emphasize their contrasting trends. The ranges of their monthly anomalies are very large, which make the long-term trends difficult to recognize. In addition, we use monthly anomalies for the correlation coefficient calculation in Figs. 6 and 7.

3) Comment: Auto-correlation analyses might have been undertaken to assess how other co-variates influence optical extinction. For example, the authors could have investigated how mean dew point influenced optical extinction, and thus assess the role of hygroscopicity on extinction for the different regions.

Response: Thanks for reviewer's nice suggestions. We searched literature and found

ACPD

12, C7391–C7397, 2012

Interactive Comment

many publications in this field, i.e., how the meteorological conditions influence PM10 or optical extinction. We cited these publications and discussed related results in places of the revised paper, i.e., Page 10, Lines 25-31.

Other Comments/Queries on material in order of appearance in the paper

1) Comment: Page (P) 17916 Line (L) 21-23 It is not evident how the correction of the impact of relative humidity on visibility was made, nor is it described in detail in the paper.

Response: We added the equations that we used for relative humidity correction (Page 4,Line 11).

2) Comment: P17916 L27 and sentences that follow 'These observations' – should 'spell out clearly' what are 'these observations'. It would seem that 'these observations' refer to 'manual' observations; yet the follow up sentences seem to infer otherwise

Response: We revised the related sentences to (Page 5, Lines 1-5): This change from manual assessments to instrument observations introduced discontinuity of the data of visibility (Wang et al., 2009). Therefore, for the U.S., we do not use the manual assessment of visibility but retained the instrument observations to provide detailed information on the relationship between visibility and optical extinction coefficient of aerosols.

3) Comment: P17919 L23-25 The number of urban, suburban and rural Canadian sites are not given for PM10 and PM2.5 – they should be specified, as it is not straightforward trying to infer numbers from Figures 1, 2

Response: We added these numbers to the main text (Page 7, Lines 13-15 and Page 8, Lines 4-5) and Table 1.

4) Comment: P17921 L15 The measurement method used for the three regions should be Described Lack of knowledge of the method(s) of measurement of PM10 for Chinese sites is deemed to be a weakness of the paper.

ACPD

12, C7391–C7397, 2012

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Interactive Discussion

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Response: The measurement methods are different site by site. It is difficult to make it easily understand except for a big table, such information can be found at the websites given in this paper. In addition, we concentrate on long-term trend rather than absolute values of the observations. The impact of the measurement methods is much less. We agree with the reviewer on the conditions of China. The related information has not been released.

5) Comment: Table 1: 'Total' is not explained and should be

Response: We added one sentence to explain this: The averages of all the available data are shown in the third column (Total).

6) Comment: Figure 6: There is no explanation given as to the increase in PM2.5 in Europe in 2005 – and which should be given

Response: We added one paragraph when discussing Fig. 6 in main text (Page 9, Line27 –Page 10, Line 2): Emission from wildfire is a major source of particles with diameters less than 2 ïĄm (Barnaba et al., 2011; Crounse et al., 2009; Reid et al., 2005). Wildland fire emissions showed large interannual variability (Giglio et al., 2006; van der Werf et al., 2006). The 2003 European summer heat wave caused the record-breaking forest fires in Portugal (Garcia-Herrera et al., 2010;Trigo et al., 2006), which contributed to the PM2.5 in 2003. Emissions of wildfires in Europe in 2003 caused wildland fire emission to be about 50 times as large as in 2008 (Rosa et al., 2011).

7) Comment: P17923 L3 'finer particles, estimated from visibility measurements' This requires clarification. Firstly, what property of fine particles is estimated ?, and secondly, on what basis is this property estimated?

Response: This sentence was in wrong places, and we deleted this sentence from the revised paper.

8) Comment: P17923 L3-4 'finer particles have an opposite long-term trend to that of PM10' This statement needs both clarification and explanation – Figures 4 and 6 show

ACPD

12, C7391–C7397, 2012

Interactive Comment

both PM10 and finer particles (aka PM2.5) having declining or negative trends over the longer term

Response: This sentence was in wrong places, and we deleted this sentence from the revised paper.

9) Comment: P17923 L12-13 It is not made clear why 'the largest seasonal variations' of both PM10 and optical extinction necessarily leads to the 'strongest' correlations

Response: We revised the paragraph to (Page 10, Lines 19-24): Monthly PM10 and optical extinction were closely correlated (Figs. 7 and 8). Both PM10 and optical extinction have the largest seasonal variations in China. The correlation coefficients between PM10 and optical extinction are the strongest in China. The concentrations of PM10 in the U.S. and in Canada are very low as is also optical extinction and both have low seasonal variations (Table 1) and much lower correlation coefficients, impacted by the measurement uncertainty.

The last sentence was added to address the reviewer's comment.

10) Comment: P17924 L1-3 Extinction coefficient derived from satellite inferred aerosol optical depth (AOD) is a column integrated value over the whole atmospheric vertical column. The inference of aerosol extinction very close to ground level from AOD clearly requires an assumption of vertical path homogeneity – which may not indeed be the case and so its comparison with surface based aerosol optical extinction is not in general directly comparable and may well be in error. In addition, the satellite signal is over a much shorter time scale than a 24 hourly averaged surface extinction value

Response: We added one paragraph to address the reviewer's comment (Page 11, Lines 7-14): Measures of aerosols reported here (PM2.5, PM10, optical extinction) are near surface measurements. Other widely used measures of aerosols are column total, i.e., aerosol optical depth derived from satellite observations. Information on aerosol profile and atmospheric mixing layer height is needed to relate these surface measures

ACPD

12, C7391–C7397, 2012

Interactive Comment

to column total measures (Schaap et al., 2009), which may have substantial seasonal variations. However, their inter-annual variation should be very small. Therefore, it is reasonable to compare long-term trends of these surface measures to column total measures under the assumption that the inter-annual variation of aerosol profile and atmospheric mixing layer height is ignorable.

11) Comment: P17924 Section 3.4 The data shows relatively high correlation coefficients between extinction coefficient and dry day occurrences for China and Europe but much poorer correlation for the US and Canada. Why? Indeed Figure 9 for the US, shows for years 1999 and 2000, no discernible effect on extinction or PM10, despite a relatively large increase in number of occurrences of dry days.

Response: We explained this in the revised paper (Page 12, Lines 13-19): The inter-annual variations of optical extinction over China and Europe agree well with the occurrence of dry days, with averaged correlation coefficients of 0.93 and 0.66, partly because both aerosols and dry days have large inter-annual variability (Fig.10). Dry days vary less in the U.S. and Canada (Fig. 10) and their contribution to the variation of aerosols is expected to be small. Inter-annual variation of optical extinction is consistent with the occurrence of dry days in the U.S. However, the correlation between PM2.5 and number of dry days is higher in Canada (Fig. 10).

12) Comment: As mentioned above, similar type analyses for PM2.5 should be carried out with respect to extinction and dry day occurrences, as presented in Figures 9 and 10

Response: We added and discussed PM2.5 to Figures 9 and 10 in the revised paper.

Other comments 1) Comment: A discussion of limitations and consequences of the use of a daily average of visibility, as pointed out by Husar et al (2000), are lacking in the paper, and at least should be commented upon.

Response: We agree with the reviewer on this. When the works are done, we only

ACPD

12, C7391–C7397, 2012

Interactive Comment

have daily averaged observations for global study. Our previous experience shows that daily visibility also works well for this kind of this study (Wang et al., 2009).

2) Comment: Attempting to seek a correlation between mass (proportional to aerosol particle diameter cubed) and optical extinction coefficient (proportional to diameter squared and dependent on particle size distribution and refractive index) is problematic. One may of course obtain a good correlation between mass and extinction for a given location for a particular airmass in which the aerosol particle characteristics do not vary greatly with time. However, there is no sound physical basis to expect a strong correlation between particle mass and particle extinction coefficient and correlations found in the paper are arguably fortuitous.

Response: Optical extinction and mass concentrations are two widely used measures of atmospheric aerosols. Previous studies have shown good relationship between these two measures at short time scales, i.e., from hourly to monthly as meteorological conditions dominate variations in these time scale. This paper shows that the long-term trend for these two measures may be very different as they are determined by different emissions. PM10 has been routinely measured and been released to the public. This paper help to correct public perception of long-term variation of aerosols based on another conventional observation, meteorological visibility.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 17913, 2012.