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Atmospheric Chemistry and Physics Discussions

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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General Comments 1) Comment: The paper collates and compares recent trends over 1-2 decades in near-surface airborne particles for 4 regions: China, Europe, US and Canada. Trends are evaluated from air-quality and visibility measurements, and are presented for three main particle metrics: (i) the mass concentration of particles with effective aerodynamic diameters <10um (PM10), based on air quality measurements; (ii) the mass concentration of particles with effective aerodynamic diameters <2.5um (PM2.5), also based on air quality measurements; (iii) amounts of optical extinction, which are linked to the abundances of particles with effective aerodynamic diameters

<1.0um and are based on visibility measurements.

The main research question being addressed may be posed as: “Are recent regional trends in near-surface PM10 paralleled by trends in finer particles like PM2.5 (which is a more important particle size for human health) and by trends in visibility that can be linked to PM1 (which is a more important particle size for radiative climate) ?” Trends are evaluated separately for urban, suburban and rural land-use situations, although data are not available for all these situations in some regions. The results for PM10 mass concentrations [metric (i) above] show a general decline of a few 10s of percentage points in China, Europe and the US, and a lesser decline of about 10% in Canada. The results for PM2.5 mass concentrations [metric (ii)above] show a general decline of a few 10s of percentage points in the US and a similar but more variable decline in Europe, but little overall trend in Canada; there are no PM2.5 data for China. The results for the optical extinction [metric (iii) above] show only a marginal (5%) decrease in Europe, and upward trends of about 10% in China, US and Canada. The authors conclude that the mass-based concentrations of the larger particles [metrics (i) and (ii)] have tended to decline in all regions over the last 1-2 decades, but that the extinction-based amounts of finer particles [metric (iii)] have tended to increase. In discussion this disparate behaviour for different particle metrics is tentatively attributed to a difference between the effects of (a) emission controls for larger particles like PM10 (e.g. controls on power plant emissions) and (b) increased emissions of finer anthropogenic aerosols (e.g. derived from vehicle exhaust gases). The paper collates and considers a wide variety of aerometric data over different geographic regions, and over time periods that overlap but that are not entirely consistent. It is difficult to audit the trend results with full confidence because of (a) the diversity of data types, (b) the variety of monitored land-use situations and time periods, and (c) limited information on measurement techniques and uncertainties. For similar reasons it is difficult to conduct rigorous statistical tests, so that the identified trends appear to be indicative rather than definitive. Nonetheless, the conclusion that a mostly upward trend in a finer particle metric [(iii)] has contrasted with a general decline in two larger particle metrics [(i) and (ii)] appears

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to be reasonably robust.

Response: Thanks your recommendation on our work. Please find our point-to-point response to your comments and suggestions.

2) Comment: One of the challenges with this type of analysis is to summarise clearly the various types and availabilities of data, and the techniques used for collating them – including the techniques for checking or correcting them. In order to help readers keep track, it may be useful to have a table that summarises data availability and collation techniques for each region, particle metric, land-use situation and period. This table could perhaps also indicate or comment on data limitations in each region. China is a key region for this study, and it is unfortunate that there is major uncertainty regarding the methods used to measure PM10 at Chinese sites.

Response: Data availability of PM10 and PM2.5 can be found at Table 1. Visibility data are available for all the regions and cover the whole study period. A summary of land-use situations has also been added into table 1.

We find the many measurement methods are used to each region and there are no specific rules for the usage of measurement techniques. To do so, a long table is needed, such information can be found at the websites given in the paper. General reader may have no interest on this. We added a paragraph into Section 2.4 that focuses on data consistency to explain this (Page 8, Lines 8-11): Many measurement techniques have been used to measure PM10 and PM2.5. The measurement technique to measure PM10 and PM2.5 at each station can be found at the above mentioned websites except for those in China. Inter-comparison of the measurement can be found at Allen et al. (1997), Price et al. (2003), and Williams et al. (2000).

3) Comment: There could usefully be more discussion of other uncertainties or alternative interpretations of the causes of particle trends, for example: Have the local land-use situations of some air-quality monitoring stations changed over the ∼20-year study period (e.g. because of urban expansion into rural areas), and have any such

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changes been taken into account ? Can we be confident that forest fire events have been sufficiently extensive in time and space to impact on some long-term regional trends, as is suggested ? âĂć Could the trends be perturbed by ENSO cycles that may induce more forest fires or photochemical pollution in some regions at particular phases of the ENSO cycle ?

Response: Thanks. For the first question, we searched literature and documents with the data. We found Andrews (2008) reported that changes of measurement location of PM10 in Beijing (Page 8, Lines 26-28). We did not find further information on other stations.

For the second question, we added a paragraph to discuss this (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).

We deleted the discussion on impact of wildfire on PM10 in the U.S. In the revised paper, we focus on impact of wildfire impact on PM2.5.

For the third question, we added a paragraph to discuss this (Page 10, Lines 25-31): High correlation coefficients between monthly PM10 and optical extinction occur because atmospheric aerosols (Mahowald et al., 2007; van Oldenborgh et al., 2010) and PM10 (Barmpadimos et al., 2011) are predominately determined by meteorological conditions at this time scale. However, long-term trends of atmospheric aerosols are little impacted by changing atmospheric circulation (Chiacchio et al., 2011; Hirdman et al., 2010; van Oldenborgh et al., 2010) but rather are dominated by emissions (Fenger, 2009; Murphy et al., 2011).

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4) Comment: The possible confounding effect of changes in the number of dry days on trends is considered for the optical extinction metric [(iii)] but not for the PM10 metric [(i)] or for the PM2.5 metric [(ii)]. Such selective consideration of confounding factors detracts from the robustness of the conclusions drawn, and it would be better if the effect of dry day changes was considered for all 3 metrics.

Response: Following reviewer's comment, we added information of PM2.5 to Figs 9 and 10.

5) Comment: The paper focuses on land-use situations for which there are reasonably large numbers of observing stations – for both air quality and visibility. Consequently, only 3 land-use situations are covered: urban, suburban and rural. Consideration of remote situations may not be possible because of a paucity of remote observing stations; neverthelss, the paper would perhaps be strengthened by some reference to published trends at remote stations, in order to compare with the trends identified at urban, suburban and rural stations ?

Response: Following the reviewer's comment, we added one paragraph (Page 10, Lines 9-11): Most of data used in this study are located in urban areas. The decreasing trends of PM2.5 reported here are consistent those at remote sites in Europe (Barmpadimos et al., 2011; Barmpadimos et al., 2012; Cusack et al., 2012), and in the U.S. (Murphy et al., 2011).

6) Comment: The authors deserve credit for attempting to identify signals of regional atmospheric particle changes from such extensive and diverse data. This kind of exploratory and retrospective analysis is limited by the amounts and types of measurements available; it is also necessarily opportunistic because the measurement networks were not originally designed for the purpose of regional trend comparisons. The results seem to constrain estimates of changes in regional particulates and aerosols, and so they should be of interest for comparison with other estimates of atmospheric particle change e.g. estimates from atmospheric modeling of regional emissions, trans-

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port and chemistry.

Response: Thanks for reviewer's recommendation. We added one paragraph to summarize the trend comparison in the context of other estimates, including satellite retrievals and model simulations (Page 14, Lines 3-15): Providing accurate estimates of long-term trends of global atmospheric aerosols is difficult. Satellite provides retrievals of atmospheric aerosols. However, these trends may be very different between satellites. For example, trends derived from MODIS (Moderate Resolution Imaging Spectroradiometer) substantially differ from those from MISR (Multi-angle Imaging SpectroRadiometer) (Zhang and Reid, 2010) and AVHRR (Advanced Very High Resolution Radiometer) (Zhao et al., 2008). Besides shortcomings of the algorithms, small satellite sensor calibration uncertainties of <2% can lead to spurious conclusions about long-term aerosol trends (Levy et al., 2010). For example, Terra's global AOD bias changed with time, so it gave an overestimation (by $\sim$0.005) before 2004, but an underestimation by a similar magnitude after (Levy et al., 2010). The year-to-year variability of aerosols is generally underestimated by state-of-the-art chemistry and transport models (Colette et al., 2011) and more should be done with these models to obtain accurate estimates of long-term variation of atmospheric aerosols (Kukkonen et al., 2012).

Specific comments (in order of appearance in paper) 1) Comment: Introduction. It would be helpful to explain that air quality monitors with size-selective inlets are designed to sample particles with aerodynamic diameters up to the nominal diameter. It follows that particles measured with a PM2.5 inlet are a subset of particles measured with a PM10 inlet. Thus there is necessarily some overlap between particle measurements of PM10 and PM2.5. Similarly, measurements of PM10 and PM2.5 should include particles <1um – which are linked to optical extinction data (visibility measurements) – so there is also some overlap between the measurements of (a) PM10 and PM2.5 and (b) optical extinction. It would be helpful to have more discussion of the extent to which the 3 particle metrics are overlapping or discrete measures of particle abundance. The relationship between optical extinction and particle size is a key con-

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sideration in the paper, and it would be helpful to have more details of this relationship. The term “fine particles” and the comparative term “finer particles” are used at several places in the paper, but it is not always clear which of the 3 metric(s) [(i), (ii) or (iii)] are being referred to on each occasion. For example, is the term “fine particles” is applied to PM2.5 and optical extinction aerosols, in order to distinguish them from PM10 ? It would be helpful to check that these terms are used clearly and consistently on each occasion. It may be helpful to explain that the PM10 metric is more indicative of near-field primary emissions of particulates, whereas the optical extinction metric is more indicative of regional-scale secondary particles (aerosols) formed over a few days and involving more atmospheric transport and chemistry. It may also help to explain that the PM2.5 metric is somewhat intermediate between these two cases e.g. it includes appreciable components of both primary and secondary particles.

Response: Thanks for these great suggestions. To keep the flow of Introduction, we added a paragraph to Section 2 (First paragraph, Page 3, Lines 11-21): PM10 is more indicative of near-field primary emissions of particulates, whereas the optical extinction is more indicative of regional-scale secondary particles formed over a few days and more involving atmospheric transport and chemistry. PM2.5 is somewhat intermediate between these two cases, i.e., it includes appreciable components of both primary and secondary particles (Fenger, 2009). PM10 and PM2.5 are designed to sample particles with aerodynamic diameters up to the nominal diameter. For example, particles measured by PM2.5 are a subset of particles measured by PM10. Thus there is some overlap between particle measurements of PM10 and PM2.5. Similarly, measurements of PM10 and PM2.5 should include particles less than 1 ïĄ■m – which are linked to optical extinction data (visibility measurements) – so there is also some overlap between the measurements of (a) PM10 and PM2.5 and (b) optical extinction.

We checked the usage of “fine particles” and tried to define it when its meaning is unclear.

2) Comment: 17916 ll4-6. Line 6 states “their optical extinction” which (presumably)

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alludes to PM10 and PM2.5 at Line 4 in the previous sentence. However, 17915 ll 13-15 links “optical extinction” to PM<1, rather than to PM10 or to PM2.5. There therefore appears to be some inconsistency between pp 17915 and 17916, because in the former page “optical extinction” is linked to PM2.5 and PM10, whereas in the latter page it is linked to PM<1.

Response: We revised this sentence to (Page 3, Lines 24-26): The meteorological visibility is used to characterize the climatic variability of optical extinction of aerosols, in particular, those with diameter less than 1 ïAęm.

3) Comment: 17916 ll19-23. These lines explain that the near-surface optical extinction coefficients of aerosols were corrected for (a) hydrometeors, and (b) relative humidity. It would be helpful to have some outline description of the methods and principles behind these corrections, and to have some indication of how large and extensive the corrections were.

Response: We added Equations used to do the relative humidity correction to Page 4, Line 11.

4) Comment: 17917 ll1-3. It appears that instrumental observations of visibility were retained for evaluating optical extinction coefficients in the US, but that manual assessments were used for this purpose in China, Europe and Canada. Does the adoption of different measures of visibility in (a) US and (b) China/Europe/Canada have any implications for regional trend comparisons ?

Response: This does not impact the long-term trend. The data collected by two different measurement methods have discontinuity (Page 5, Lines 1-3).

5) Comment: 17917 ll 4-18. It is not clear what types of land-use situations were covered by visibility measurements, because unlike the PM10 and PM2.5 data they are not resolved into urban, suburban and rural situations. This hinders like-for-like comparisons between extinction and PM data i.e. comparisons between data from the

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same type of land-use situation. More generally on the subject of land-use, it would be useful to know if rural sites included “remote” sites that were minimally exposed to anthropogenic influences; it would also be useful to have some discussion of other land-use considerations such as the effect of sea salt near coasts.

Response: The results shown in this paper are consistent with measurements at remote sites (or background) (Page 10, Lines 9-11): Most of data used in this study are located in urban areas. The decreasing trends of PM2.5 reported here are consistent those at remote sites in Europe (Barmpadimos et al., 2011; Barmpadimos et al., 2012; Cusack et al., 2012), and in the U.S. (Murphy et al., 2011).

We also show that aerosols derived from visibility over the China are consistent with those over ocean (Page 11, Lines 15-27): We find that the increase of optical extinction in China is consistent with the increase of SO2 (an important precursor of particles with diameter less than 1 ïA■m) estimated from satellite retrievals (Wang et al., 2010; Lyapustin et al., 2011; Itahashi et al., 2012), ground-based measurements (Lu et al., 2010), and model simulations (Lu et al., 2010). In particular, satellite-derived aerosol optical depth (figure 4a of Itahashi et al., 2012) and modeled SO2 emission (figure 2 Lu et al., 2010) show nearly same variability of optical extinction of aerosols derived from visibility. The cessation of the rapid increase since 2005 has been attributed to the application of flue-gas desulfurization devices in power plants in response to new Chinese policy (Lu et al., 2010). This flue gas desulphurization technique was used by most European, US and Canadian power stations before the start of the study period and therefore its introduction did not impact the long-term variation of aerosols in Europe, U.S. and Canada over the period studied (Cooper et al., 1997; Nan, 1994). Several of China’s mega-cites announced other measures (Wang and Chen, 2010) to control air pollution after 2005.

6) Comment: 17923 l10. Figures 7 & 8. It is interesting that the monthly variations of PM10 and optical extinction are positively correlated, but that their longer ($\sim$1-2 decade) trends appear to diverge (decreasing PM10 v. generally slightly increasing ex-

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tinction). It would be helpful to suggest processes that could account of this difference – perhaps because monthly variations are dominated by meteorological variations between months, whereas longer trends are more sensitive to trends in emissions.

Response: Following the reviewer's suggestion, we added one paragraph to explain this (Page 10, Lines 25-31): High correlation coefficients between monthly PM10 and optical extinction occur because atmospheric aerosols (Mahowald et al., 2007; van Oldenborgh et al., 2010) and PM10 (Barmpadimos et al., 2011) are predominately determined by meteorological conditions at this time scale. However, long-term trends of atmospheric aerosols are little impacted by changing atmospheric circulation (Chiacchio et al., 2011; Hirdman et al., 2010; van Oldenborgh et al., 2010) but rather are dominated by emissions (Fenger, 2009; Murphy et al., 2011).

7) Comment: 17924 ll1-2. Presumably, "satellite-derived" aerosol optical depth are estimates through the whole overlying atmosphere, so not compatible with the "near-surface" observations of visibility, PM10 and PM2.5 ?

Response: We added a paragraph to explain this (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 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.

8) Comment: 17924 ll3-7. Need to explain that flue gas desulphurization (FGD) was already fitted to most European, US and Canadian power stations before the start of the trend periods, so that, unlike in China, there is no effect during the trend periods

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from fitting FGD in these first 3 regions (?)

Response: We added one sentence to explain this (Page 11, Lines 23-26): This flue gas desulphurization technique was used by most European, US and Canadian power stations before the start of the study period and therefore its introduction did not impact the long-term variation of aerosols in Europe, U.S. and Canada over the period studied (Cooper et al., 1997; Nan, 1994).

9) Comment: 17925 l17-19. It would be helpful to discuss if you would expect flue gas desulphurization to suppress all 3 particle metrics (PM10, PM2.5 and optical extinction) and/or if it would effect them by different amounts ?

Response: We have evidence that flue gas desulphurization suppresses SO2 and optical extinction as discussed in Page 11, Lines 15-28. We don't find information on other two metrics.

10) Comment: 17926 ll6-7. This gives "more fossil fuel emission from developing countries, such as China. . ." as a proposed "reason for the increase of optical extinction". However, this should be qualified by noting that more recently there has been a decline in the rate of increase that is likely due to the application of flue gas desulphurisation in China.

Response: Thanks for pointing it out. We added one sentence to explain this (Page 13, Lines 21-22): Since 2005 in China, there has been a decline in the rate of increase that is likely due to the application of flue gas desulphurization (Lu et al., 2010).

11) Comment: 17926 ll11-13. In order to make comparison with black carbon, it would be helpful to explain how the black carbon metric compares with the metrics used for PM10, PM2.5 and optical extinction. For example, it would be helpful to explain if the black carbon metric is most associated with larger or smaller particles, and how it relates to mass-based and visibility-based metrics.

Response: We revised the sentence to address the reviewer's comment (Page 13,

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Lines 28-31): A recent study consistent with our results shows that the relative increase in anthropogenic black carbon that has a particle size less than 1 ïĄm and is efficient in optical extinction, is much larger than the overall increase in the anthropogenic abundance of aerosols (Myhre, 2009).

Technical Corrections (in order of appearance in paper) 1) Comment: 17916 l27. The statement “These observations are not homogeneous” could be understood to refer to the aforementioned “instrumental (visibility meter) observations”, but not to the “manual assessment of visibility”. For clarity , it would be better to say instead “These two measures of visibility are not homogeneous”.

Response: We deleted this sentence and 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.

2) Comment: 17921 ll24-25. The comment that “urban areas in European countries and the US are lightly polluted in terms of PM10” is apparently intended to draw a contrast with more heavily polluted urban areas in China (?). If so, it would be better to say so explicitly: “urban areas in European countries and the US are lightly polluted in terms of PM10 compared to urban areas in China”.

Response: Done

3) Comment: 17922 l25. This says ‘The duration of the PM2.5 data is shorter than that of PM10”. Actually the PM2.5 series are not much shorter i.e. 12-15 years duration compared with 12-18 years for PM10 (?). It would be better to say “The duration of the PM2.5 data is slightly shorter. . .” ?

Response: Done.

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4) Comment: 17924 l25. For “the agreement of PM10 a number of dry days is better in Canada” substitute “the correlation between PM10 and number of dry days is higher in Canada”.

Response: Done.

5) Comment: Fig. 7 Caption. Change 3rd line to read: “column: same as left column except that correlation coefficients are between monthly anomalies”.

Response: Done.

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