

Review of 'Contrasting Trends of Mass and Optical Properties of Aerosols over the Northern Hemisphere from 1992 to 2011' by K.Wang, R.E.Dickinson, L.Su, and K.E.Trenberth.

General Comments

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 to be reasonably robust.

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.

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 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 ?
- 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 ?

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.

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; nevertheless, 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 ?

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, transport and chemistry.

Specific comments (in order of appearance in paper)

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 <1µm – 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 consideration 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.

17916 II4-6. Line 6 states “their optical extinction” which (presumably) alludes to PM10 and PM2.5 at Line 4 in the previous sentence. However, 17915 II 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.

17916 II19-23. 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.

17917 II1-3. 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 ?

17917 II 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 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.

17923 I10. Figures 7 & 8. It is interesting that the monthly variations of PM10 and optical extinction are positively correlated, but that their longer (~1-2 decade) trends appear to diverge (decreasing PM10 v. generally slightly increasing extinction). 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.

17924 II1-2. 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 ?

17924 II3-7. 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 from fitting FGD in these first 3 regions (?)

17925 I17-19. 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 ?

17926 II6-7. 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.

17926 II11-13. 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.

Technical Corrections (in order of appearance in paper)

17916 I27. 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*”.

17921 II24-25. 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”.

17922 I25. 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...” ?

17924 I25. 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”.

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