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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Monthly-averaged anthropogenic aerosol direct radiative forcing over the Mediterranean from AERONET derived aerosol properties" *by* A. Bergamo et al.

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General Comment

This papers presents an interesting estimate of the direct radiative forcing by anthropogenic aerosols at land and coastal sites throughout Italy from 35.5 to 45.8°N plus Crete. It is based on the combination of a nice data set derived from six AERONET photometer monitoring stations operated in 2003, model estimates of the fraction of anthropogenic aerosols, MODIS estimates of the surface albedo, a cloud climatology to take into account cloud coverage, and a radiative transfer model. The paper first presents optical properties for the total aerosol from Aeronet data and additional Mie

computations, and then focuses on the submicron fraction to derive radiative forcing. I find the approach interesting, most results are discussed and illustrated in detail, and the bibliography is good. A number of minor points, typing errors, etc, have been clarified or corrected by the authors following my initial technical review. In my view the paper brings new information quite relevant to ACP, but there are points that deserve to be clarified or improved. I detail in the following my specific comments and suggestions for reviewing the paper.

Specific Comments

1. I regret that it is not briefly investigated (i) how representative of the Central Mediterranean are the AERONET sites used and (ii) how variable is the aerosol in the given region of interest from one year to the other. In 2003, March was exceptionally polluted in North Italy as said in the manuscript. Also August experienced large forest fires and a heat wave in southwestern Europe which caused high loads of submicron aerosols (e.g. Pace et al., JGR, 2005). You might comment at some point on how the August heat wave and large fires in southern Europe translate into aerosol optical properties (see top of p 12785, or peak in atmospheric heating in August in Fig. 12). But by the way, such a specificity of year 2003 implies that radiative forcing by anthropogenic aerosols may be somewhat larger than on average because of higher anthropogenic AODs than normal. At least a brief warning would be necessary. MODIS and/or other satellite data (see e.g. the 7-yr Mediterranean aerosol climatology by Antoine and Nobileau , J. Geophys. Res., 111, doi:10.1029/2005JD006795, 2006) could be used to comment on this interannual variability.

2. I have some concern with the meaning of the present overall averaged radiative forcing. Five of the six stations have surface albedos more or less representative of coastal areas (depending on their position relative to the MODIS 1° grid) and we have no sensitivity study to the influence of the surface albedo. I consider unwise making the 6-station average and concluding that it is representative of the forcing for Mediterranean land sites. I would rather stay on an overall range from the various stations.

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3. The radiative forcing is computed here for land and coastal sites with relatively high surface reflectances compared to the open sea. The lower surface albedo of the open sea is expected to yield somewhat different radiative forcing in the solar spectrum. This also implies that present results are unfortunately not applicable to a major part of the Mediterranean. In order to check the surface albedo effect on the radiative forcing and to derive the solar forcing over marine areas off the coastal stations, I suggest that radiative forcing is additionally computed using open sea surface albedo (assuming that aerosol properties can be extrapolated off-shore). This could be done at least for one of the southern stations. If done more systematically for the coastal sites, it might give more sense to averaging results into one number which could be more representative for the Central Mediterranean region than the present average (see point 2).

4. There is no indication on uncertainty/variability in surface albedo and no sensitivity study to this parameter, although it is one of the most influent in radiative forcing calculations. Since there are two monthly values from MODIS and that only the monthy average is used in radiative computations, information on these two values might be plotted in Fig. 2 and a sensitivity test might be performed.

5. Given the detailed data set available from AERONET, information on the daily variability would be useful. This is why I suggest to plot std deviations (based on the daily variability) for each monthly average in Figs. 3-5. Since this will add much information on the plots, I would split those figures into 3 sub-plots rather than 2, with the stations grouped by latitude band (Ispra/Venice, Lecce/Oristano, and Lampedusa/Crete) since this latitude parameter seems to explain most of the variability.

6. The paper is relatively long and the methodological scheme is rather complex. Confusion is easy between measured and computed parameters on one side, or between values for total and anthropogenic-only aerosols on the other. I think it is necessary to help the reader by including a diagram showing the methodological scheme and flowchart with the various models used, the input parameters and their origin, the outputs, and possibly respective tables and figures where to find which values.

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7. In particular there is some ambiguity coming from the fact that you seem to perform Mie computations to compute optical properties, firstly for the total aerosol and secondly for the anthropogenic (submicron) fraction only (worth confirming that only the size distribution changes compared to preceding section).Only the first results are deeply discussed and illustrated in Figs 3-5, whereas the last results are further used to compute radiative forcing but are not shown. How different are they from those of the total aerosol? As done in other papers (e.g. Yu et al., 2005) and for traceability of final results, it would be worth to compare the optical properties of the total aerosol and of the submicron fraction.

8. At the beginning of page 12774, it would be worth briefly explaining the selection criteria of the "selected AERONET sites".

9. About AERONET products in Section 2:

9.1. Direct sun measurements can accommodate a low cloud coverage, but sky radiance measurements cannot. So it would be worth to recall that AERONET measurements are essentially clear-sky and that it is assumed in this work that the average aerosol properties derived from AERONET can be extrapolated to all sky conditions.

9.2. I guess that you obtain from AERONET values for eta, the ratio of the accumulation mode fraction AOD to total AOD. This is worth to mention in p. 12773. Note that this is a product of level 1.5 or 1, not 2.

9.3. It is necessary to specify whether the level 2 AERONET particle size distribution product used here takes into account particle non-sphericity, because desert dust (non-spherical) is an important component of the Mediterranean aerosol: not taking into account the presence of non-spherical particles in the retrieval of the aerosol distribution in the presence of dust in the AERONET inversion yields a strong bias in the accumulation-mode fraction of the particle size distribution (Dubovik et al., Geophys. Res. Lett., 29(10), doi: 10.1029/2001GL014506, 2002).

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9.4. It should also be specified at end of page 12773 that a significant number of level-2 data are not available for the refractive index so that level 1.5 data are considered for those days (as a consequence replace "It is worth noting" by "We recall" in line 19 of p. 12776).

10. I would expect some numbers on refractive indices and size distributions are added. Quantitative information on such quantities will provide additional useful data, and is needed to the traceability of final results

11. Because AERONET retrieves an "average" refractive index, which is in reality different between the coarse and the submicronic particle size fractions, and because dust is often present over the Mediterranean (e.g. Antoine and Nobileau, 2006), I suggest to look for extreme values of refractive indices observed during only high pollution episodes, and check whether their variability is not without the range used for the sensitivity study.

12. I have a concern with the forcing calculations in the infrared. Especially, I do not understand the point on refractive indices in the infrared (p. 12776, lines 12-15). Why should anthropogenic submicron particles be given refractive indices of dust based on the fact that only dust particles have a substantial effect in the infrared? My understanding is that the dust interaction in the infrared is mainly due to its coarse size distribution, implying scattering, (e.g. Dufresnes et al., Longwave Scattering Effects of Mineral Aerosols, J. Atmos. Sci., 59, 1959-1966, 2002). The submicron fraction is too small to scatter thermal infrared significantly. So I guess that the radiative impact of anthropogenic aerosols found in the infrared stems from absorption. Dust particles have some absorption in limited spectral regions that are quite variable depending on mineral types. Are absorption bands of quartz, calcite, clay, etc, really applicable to anthropogenic submicron particles? What kind of absorption is supposed in this work for anthropogenic particles needs to be clarified.

13. One additional reason for plotting std deviations in Figs. 3-5 is that for certain

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months at several stations, the monthly averaged parameters rely on very few days. This lack of measurements likely causes some of the "noise" in the monthly time series, and std dev. bars could help in interpreting the reality of month to month variations. For instance at Oristano in October 2003 a high desert dust episode strongly affects the monthly mean values based on 3 days. The December month has only 1 d of measurements in Crete. Etc. Even if this has not a great impact on the final product of yearly averaged radiative forcing, I think that a warning with a reference to table 1 is necessary in the figure legends on optical properties, so that the readers eventually reuse cautiously some of the monthly means plotted. I suggest that some contrast is given in the figures by using specific filled or open symbols in every station series in order to highlight those months where only 1 to 3 days (i.e. less than 10%) are available.

14. Aerosol vertical profile (section 3.4):

14.1. Significant seasonal variations are expected in the height of the atmospheric boundary layer (see e.g. a paper by Dayan in Guerzoni and Chester book The Impact of Desert Dust Across the Mediterranean, Kluwer, 1996) and Guibert et al. also report seasonal variations in aerosol vertical profiles. It should be stated that seasonal variations are neglected.

14.2. Furthermore Guibert et al. show that the vertical profiles of aerosol extinction in southern Europe are significantly affected by dust. If the profile used here really stems from their average southern European profile of aerosol extinction, it is not adapted to anthropogenic aerosols. It would be better considering the profile they compute for anthropogenic aerosols only.

14.3. I do not understand the x-axis legend in Figure 6. The "normalized AOD" quantity seems uncorrect: a constant AOD between the surface and 0.5 km altitude means that there is no aerosol within this layer. I think the x-axis should rather read "Relative aerosol extinction" (or aerosol concentration?) and be normalized between 0 in the

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free troposphere and 1 at the surface (to be specified in the figure legend).

15. Cloud cover (section 3.5): is 2003 a normal year in terms of cloud coverage compared to the decadal climatology?

16. I find that the discussion, at bottom of p. 12780, of aerosol types depending on the value of eta (ref. to Bellouin et al.) would be better placed in p. 12778 when discussing the values found at the different stations.

17: Is there any relationship between the seasonal cycle in SSA or g and relative humidity?

18. Top of p. 12785: at some southern stations, likely due to forest fires in Portugal (Pace et al., 2005), SSA is also relatively low in August, which contributes to higher forcing.

19. I would add a line with total AOD on top of table 3. If my computations are right, Crete has a greater AOD than Oristano, and Lecce has a greater AOD than Venice: this somewhat contradicts conclusions on top of page 12792.

20. Fig. 11-12: You might comment why a minimum is found in July in both TOA AFE (Fig. 11) and SFC AFE (Fig. 12) at all stations except at Oristano and Venice, where the July minimum in TOA AFE is particularly marked (Fig. 11) and where the minimum in SFC AFE is in August (Fig. 12).

21. I think that some references could allow an interesting comparison with regional radiative forcing from anthropogenic greenhouse gases.

Technical comments:

-Line 14-15, p. 12776: can "only" be attributed...

-End of page 12776: the section on MIE calculations (lines 22-28) is misplaced here because it introduces new variables which are not necessary in the following long discussion on Figure 3. It would be better to move it in page 12778 (line 17) where relevant

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variables are presented in Figs 4 and 5.

-Line 1, p. 12777: monthly averaged "measured" AODs...

-Line 7, p. 12777: and the accumulation, "in the absence of rain", of local...

-Line 18, p. 12777: total optical depth " (at 550 nm) ".

-Line 19, p. 12777: eta "is used to infer" rather than "represents the best parameter"; refs can be given here.

-Line 12, p. 12777: outbreak "that" occurred...

-Line 14, p. 12777: Figure 3b show"s" that...

-Line 20, p. 12779: ref. to Zhou et al. at end of sentence is strange; what does it bring compared to Yu et al. cited earlier? Note that Fig. 4 in Yu et al. rather reads 0.66 for the total aerosol (or 0.64 for the fine mode) than 0.65 as stated.

-Line 2, p. 12783: statistic"s"...

-Line 5, p. 12783: interpolated "from"...

-Line 24, p. 12787: several "other" chemical transport models...

-Legends of Figs 4 and 5: they seem to apply to the total aerosol. A note might specify this more clearly since the focus of the paper is the anthropogenic (fine mode) fraction.

-Table 1: I suggest to add a "Total" right column and bottom line.

-Tables 3 and 4: Clarify what the mean, min-max and ś do refer to: monthly means?

-Table 6 to 9 might probably be grouped into a single table.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 12769, 2008.

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