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## Interactive comment on "Significant variations of trace gas composition and aerosol properties at Mt. Cimone during air mass transport from North Africa – contributions from wildfire emissions and mineral dust" by P. Cristofanelli et al.

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**General Remarks** "The manuscript describes an important issue, the mixing of Saharan dust with other types of polluted air masses that is not easily accessible experimentally. The method of analysis looks sound. The interesting approach based on age analyses should be stressed somewhat more in the Abstract and the Conclusions. Also the role of biomass burning to the future development of the Mediterranean atmosphere is an important aspect that could yield value to this study. The manuscript is clearly written, cites a lot of relevant literature, and should be published after consider-

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## ing a few changes suggested in the following."

The authors thank the referee for the valuable suggestions that will further improve the paper. In the following our answer to each specific remarks are listed.

## **Specific Comments:**

1)Abstract: Add a few words about the analysis of the air-mass age.

Also following the suggestions of the referee 2 and 3, the abstract will be changed as following: "High levels of trace gas (O3 and CO) and aerosol (BC, fine and coarse particles) concentrations, as well as high scattering coefficient ( $\sigma$ s) values, were recorded at the regional GAW-WMO station of Mt. Cimone (CMN, 2165 m a.s.l., Italy) during the period 26 - 30 August 2007. Analysis of air-mass circulation, aerosol chemical characterization and trace gas and aerosol emission ratios (ERs), showed that high O3 and aerosol levels were likely linked to (i) the transport of anthropogenic pollution from northern Italy, and (ii) the advection of air masses rich in mineral dust and biomass burning (BB) products from North Africa. In particular, during the advection of air masses from North Africa, the CO and aerosol levels (CO :175 ppbv, BC: 1015 ng/m3, fine particle: 83.8 cm-3,  $\sigma$ s: 84.5 Mm-1) were even higher than during the pollution event (CO: 138 ppbv, BC: 733 ng/m3, fine particles: 41.5 cm-3, *s*: 44.9 Mm-1). Moreover, despite the presence of mineral dust able to affect significantly the O3 concentration, the analysis of ERs showed that the BB event represented an efficient source of fine aerosol particles (e.g. BC), but also of the O3 recorded at CMN. In particular, the calculated O3/CO ERs (0.10 - 0.17 ppbv/ppbv) were in the range of values defined in literature for relatively aged (2-4 days) BB plumes and suggested significant photochemical O3 production during the air-mass transport. Moreover, for fine particle concentration and  $\sigma$ s, the calculated ERs was higher in the BB plumes that during the anthropogenic pollution events, stressing the importance of the identified BB event as a source of atmospheric aerosol able to affect the atmospheric radiation budget. These results suggest that events of mineral dust mobilization and wildfire emissions over

North Africa could significantly influence radiative properties (as deduced from  $\sigma$ s and BC observations at CMN) and air quality over the Mediterranean basin and northern Italy."

2)P. 4, lines 5-15: The role of O3 is well known. The authors should introduce this part more in view of the aspects of the air-mass characterization, perhaps condense the more general statements into one or two sentences (including the references).

In the revised paper, the sentences about ozone will be changed as following (also the number of references will be reduced following the referee 3 suggestions): "O3 is strongly involved in photochemical reactions (Crutzen et al., 1999; Volz-Thomas et al., 2002) and in determining the oxidation capacity of the troposphere (Gauss et al., 2003). Moreover it is a dangerous pollutant (Hoek et al., 1993; Brunekreef and Holgate, 2002; Conti et al., 2005; Paoletti et al., 2006) and an efficient greenhouse gas (Forster et al., 2007). Due to chemical and photo-chemical processes and mixing with air-masses of different origin, the O3 levels in the BB plumes can strongly vary during the export from the emission regions (Real et al., 2007)".

*3)P.4, text on BC (or following paragraph): Add a statement on the growing number of fires; I do not know any related literature on the Mediterranean regions, but this has already become obvious from the North American fire statistics.* 

The following statement about the growing number of fires will be added to the revised paper. "Even if year-to-year variability affect global fire activity and thus wildfire emissions (van der Werf et al., 2006; Le Page et al., 2008), significant increasing trend in fire activity were observed during the recent decades over specific regions like Europe, Africa and US (Mouillot and Fields, 2005; Westerling et al. 2006)."

Le Page, Y., Pereira, J. M. C., Trigo, R., da Camara, C., Oom, D., and Mota, B.: Global fire activity patterns (1996–2006) and climatic influence: an analysis using the World Fire Atlas, Atmos. Chem. Phys., 8, 1911–1924, 2008

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Muoillot, F., and Field, C. B.: Fire history and the global carbon budget: a fire history reconstruction for the 20th century, Glob. Chang. Biol., 11, 398–420, 2005.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano, A. F.: Interannual variability in global biomass burning emissions from 1997 to 2004, Atmos. Chem. Phys., 6, 3423–3441, 2006.

Westerling, A.L., H.G. Hidalgo, D.R. Cayan, and T.W. Swetnam: Warming and earlier spring increase western U.S. forest wildfire activity. Science, 313, 940–943, 2006.

4)P. 5, line 9: Add the recent paper Papayannis et al., J. Geophys. Res. 113 (2008), D10204; doi: 10.1029/2007JD009028

The reference will be added.

5)P. 13, line 23: (Henne et al., 2004): The orographic wind system has been studied for several decades. There have been numerous important papers on this subject. I suggest to cite at least the classical paper by Vergeiner and Dreiseitl (Meteorol. Atmos. Phys. 36 (1987), 264-286) in addition (possibly also others).

As suggested, more references will be added (Vergeiner and Dreisitl, 1987; Zaveri et al., 1995).

6)P. 14, lines 2-3: The importance of the orographic venting to the data selection for high-lying stations and the export to the free troposphere was also studied within the TOR and VOTALP projects (e.g., Scheel et al., J. Atmos. Chem. 28 (1997) 11-28; Carnuth et al., Tellus B 54 (2002), 163-185; Furger et al., Atmos. Environ. 34 (2000), 1395-1412; Carnuth and Trickl, Atmos. Environ. 34 (2000), 1425-1434.

As suggested. the following reference will be added: Scheel et al., 1997; Carnuth et al., 2002.

7)Discussion: Add one sentence to emphasize the importance of the age analysis.

As also following the suggestions by the referee 3, the sentence at page 7845 line 24,

will be rephrased as following: "Even if caution should be deserved in directly relating BB plume ages and O3/CO ERs for different BB plumes due to the large emission variations among different wildfires, several investigators (e.g. Pfister et al., 2006, Val Martin et al., 2006; Real et al., 2007) have proposed that the general increase of O3/CO ER with the plume ages could be the effect of the slow recycling of PAN, HNO3 and organic nitrates, which favour the photochemical formation of O3 in BB plumes by increasing the effective lifetime of NO2"

8)P. 22, lines 7-15 (and Abstract, p. 3, line 13): I would be careful to relate an ozone decrease to the presence of dust alone. Quite frequently, the air masses overpassing the Sahara desert originate over the remote Atlantic where ozone values of 20-40 ppb have been measured. These low mixing ratios frequently survive during the transport to Central Europe either via north-western Europe or (in some cases) the Mediterranean Sea.

We agree with the referee that also air-mass origin from North Atlantic can contribute in the lowering of O3 concentrations observed at CMN when air-mass rich in mineral dust reach the measurement site. For this reasons we will reformulate the sentence for better addressing this point: "Even if the low O3 mixing ratios usually observed at CMN during Saharan dust events may be also related to the chemical composition of air-masses originating from North Atlantic and North Africa (Bonasoni et al., 2004), significant heterogeneous O3 destruction can occur at the surface of dust aerosols (Hanisch and Crowley, 2003; Bauer et al., 2004), while HNO3 and NO3 depletion on dust particles can remove a fraction of O3 precursors (Zhang and Carmichel, 1999; Harrison et al., 2001)". Nevertheless, the authors note that, as shown by Bonasoni et al. (2004), during Saharan dust events the amount of O3 decrease was greater in air-masses with high mineral dust loading than in air masses coming from North Africa but without mineral dust, thus suggesting that the mineral aerosol can actually play a fundamental role in depleting O3 mixing ratios.

## **Technical comments:**

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All the residual technical comments have been accepted and fixed by authors

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 7825, 2009.