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Interactive Comment

Interactive comment on "Long term particle size distribution measurements at Mount Waliguan, a high-altitude site in inland China" *by* N. Kivekäs et al.

N. Kivekäs et al.

Received and published: 24 April 2009

Response to referee comments on manuscript: Long term particle size distribution measurements at Mount Waliguan, a high-altitude site in inland China by N. Kivekäs et al.

Anonymous Referee #1 Received and published: 21 February 2009

REFEREE: General comments This paper describes variations of number size distribution and CN concentration at Mount Waliguan, China. They measured about one and half year with some long interruptions. This paper includes analysis of diurnal and seasonal variations and relation with air trajectory.

However, the paper does not consider well about local setting of air circulation and adequately justify representativeness as an aerosol monitoring site. I feel that this site is a rural (?) inland place, rather than typical mountain site. This paper contains potentially useful data to show features of this kind of inland area, but description of the current manuscript is lacking new implication and conclusive findings. More analysis and discussion will be needed to provide meaningful new results. Combined above with other points listed below, this paper needs more work to be published in ACP.

AUTHORS: It is true that the site does not show some of the typical mountain site features, such as clear pattern between planetary boundary layer (PBL) and free troposphere (FT) air. The site is, however, certainly a high altitude site with low ambient pressure. Some high altitude measurement sites are located even lower compared to their surroundings (eg. Bonasoni et al., Sci Total Environ, 391(2-3), 2008, 252-261) The air circulation around the site is addressed better in the revised manuscript.

The novelty of the measured data derives mostly on the fact that no earlier long term aerosol size distribution measurements have been conducted within 1500 km radius from the Mount Waliguan station. There has been no information regarding the aerosol size distribution conditions in the area. Other than the general aerosol level this manuscript shows how the aerosol size distribution depends on the wind pattern, season and time of day, and discusses the factors affecting the distributions. The main new finding is that the particle number concentrations in this area are high, but are caused rather by natural and local (˜500km radius) sources than transported pollution from densely populated Eastern China.

Two new tables (1 and 4) and one new figure (1) were added to the revised manuscript to make the results more clear. Also several figures were modified for the same reason.

REFEREE: Specific comments

1 introduction The aim of this study is not clearly stated. To avoid local contamination, any rural site would be suitable to measure aerosols. Why do you use the mountain or

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high altitude site?

AUTHORS: There are three reasons why Mount Waliguan GAW station was chosen to be the site for this study. 1) As stated on the introduction, a site located on higher altitude than its surroundings offer a better view of the general aerosol conditions. The aerosol is better mixed and the site is not trapped inside nighttime inversion during cold periods. 2) A mountain top site offers the possibility to study changes between PBL air and FT air, even though no clear pattern was found in this study. 3) There was already a well established research station with necessary infrastructure and capable staff at Mount Waliguan. The earlier work published from the site can also be used to better understand the results.

REFEREE: 2 materials and methods 2.1 measurement site Topographic map should be included to show local settings about cities, relative height difference around this area, and local wind condition at this highland area. This site seems to be located at a hill in highland. Several papers on atmospheric chemistry at this site have been published (Tellus, 55B, 145-158, 2003; Wang, JGRD, 2005jd006527, 2006 etc). These papers discussing about local contamination might be useful to show site characteristics of local air situation. Combined figures of typical time variations for CN, water vapor, trace gas concentration, up wind velocity etc are also helpful to show local mixing situation.

AUTHORS: A more detailed description of the local topography (including a topographic map, Fig 1) and the air flow conditions around Mt. Waliguan is added to the revised version of the manuscript. The local mixing patterns are discussed in more detail. No trace gas data or vertical wind data were available for this study, so the conditions are described on qualitative level. The horizontal wind direction did not show a clear daily pattern as in Wang et al. (2006)

REFEREE: 2.2 instrumentation References for GAWSIS, FLEXTRA trajectories are needed.

AUTHORS: The scientific reference to FLEXTRA is included to the revised manuscript, as well as the url:s of the websites to both GAWSIS and FLEXTRA

REFEREE: 2.3 data processing Provide details of data screening. How long does one scan take?

AUTHORS: The scan length is mentioned in a more clear way in the revised manuscript.

REFEREE: Describe reasons to categorize size distribution and provide more about criteria for separating distribution type. For comparative purpose to other place, modal parameters would be suitable instead of subjective type separation used here. What is starting height for air trajectory?

AUTHORS: The separation to different distribution types was made from the whole size distribution with 28 size bins, not only from the three size fraction given in Figure 2. The text was rearranged to avoid misunderstanding in this matter. The modal parameters of the different distribution types are also given in the revised manuscript to make comparison easier. The modal parameters in the new Table 1 and Figure 2 should make it clear to what extent the distribution types are different from each other.

The trajectories are calculated to 625 hPa level, responding to altitude of approximately 3950 m above sea level. This was also added to the revised manuscript.

REFEREE: 3. Results 3.2 comparison to other measurements I feel that this site is a place of rural (?) highland because of absence of clear mountain-valley winds and high CN concentration. Rather than mountain site, comparison with Siberian data (such as Koutsenogii & Jaenicke, JAS, 25, 377-383, 1994; Paris et al., AE, 43, 1302-1309, 2009 etc) seems to be more suitable, as compared to Indian sites in this paper.

AUTHORS: Mt Waliguan station elevation is located geographically about 1500-2000 km from the sites in India and Nepal, and the same distance from the South-Siberian sites around Lake Baikal. The Mt Waliguan elevation and proximity of brown cloud is

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more similar to the stations in India. The climate and terrain at Mt Waliguan area are more similar to the Siberian stations. The vegetation at Mt Waliguan (arid grasslands) is very different from both the Indian and Siberian sites. To get a comprehensive picture of the aerosol conditions in the area the comparison to Siberian studies is added to the revised manuscript, but the comparison to Indian and Nepalese sites is not removed.

REFEREE: 3.3 temporal variation As mentioned in this section (lower part of P2061 and upper part of P2062), time variation of water vapor at this site seems to suggest diurnal mixing "within" PBL not for mixing between free-troposphere and PBL, probably because altitude difference of 600 m from the surrounding highland is not enough to climb up thick PBL at this area. More work will be needed to characterize aerosol data in relation with diurnal variation of PBL structure at this site.

AUTHORS: There was no more data available for a more detailed study on the diurnal variation of the PBL at the site. The vertical wind could not be derived from the horizontal wind, because the wind pattern was not as clear as in Wang et al. (2006). More information was added on the new particle formation events, and the variation with the events removed was also studied, leading to no clear pattern in the diurnal variation. The main conclusions remain: 1.) There was no systematic pattern between FT and PBL air observed at Mount Waliguan and 2.) The increased concentrations of nucleation mode particles during daytime come from new particle formation events.

REFEREE: 3.4 trajectory analysis Although trajectory analysis is presented, there is no conclusive finding.

AUTHORS: The conclusive finding of the trajectory analysis is that air masses coming from east to Mt Waliguan contain more and larger particles than other air masses. As the Eastern air masses typically originate from other sectors, the particles are more probably emitted by sources a few hundred km east of the station rather than in the densely populated coastal regions of China. A new table (Table 4) was added to better highlight this finding.

REFEREE: Technical corrections P2066 Brasseur et al, wrong title

AUTHORS: Corrected

Anonymous Referee #2

Received and published: 17 March 2009

REFEREE: The paper relates the continuous measurements of aerosol numbr concentration and size distribution properties performed at the station of Mt Waligan, Central China. The novelty of this set of measurements derives mostly from the geographical origin of the measurements covering an area which has never been well documented. Impact of aerosol emissions in China clearly raises the issue atmospheric composition changes over the entire region. This is really the relevant information from this paper to document with robust statistics (2 years) the variability of aerosol number and size at that height representative of the regional/synoptic atmospheric environment.

General comments

To my opinion, there are few corrections required at this stage on the manuscript although the paper remains somewhat descriptive. In the following, I suggest a few ideas to improve the paper.

My main concern relates to the statistical evaluation of the results and their potential use for deriving typical aerosol properties in this area. My feeling is that, at this stage, the paper fails identifying the main factors involved in the observed variability of both aerosol size and concentration which is identified as one of the main objectives. It is important to insure that a statistical analysis does not lead to mean values that do not reflect any real observation. I am not sure that all statistical evaluation presented in this paper are not biased by mixing different categories of observation. Results are sorted into categories but the statistical relevance of the sorting is no well discussed. Practically, we do not know if the observed variability is driven by day/night changes, air mass origin, seasonality (etc..) while this should be the main information

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from the paper. This is resulting from the fact that statistical approaches to categorize the variability are performed independently from each other and not organized into a hierarchy. For example, Figure 5 shows quite high diurnal variability for the nucleation mode aerosol. The variability is more marked during summer than during winter (the Figure is difficult to read on a log-scale). This is certainly related to local phenomena as discussed in the text. Now, how is this variability impacting on the Table 1, i.e. to which extent seasonal averages are biased by seasonal changes of diurnal variability (results from Table 2)? A similar problem can be raised for the representativity of the observed distributions (Fig 1), i.e. to which extent distribution types contain changes in diurnal variability. This is why a more careful hierarchy in applying statistical data reduction may lead to a more robust description of aerosol properties. My suggestion is therefore to remove the local effect seen in Figure 5 before sorting according to another variable. This can be done selecting periods during which the local influence is limited (i.e. night/morning).

AUTHORS: The seasonal pattern was re-analyzed with the periods of dominant nucleation mode removed, as well as with only the nighttime (00:00 – 08:00) values included. Neither one of these changes affected the seasonal pattern significantly. This was expected, as a large fraction of the particles produced in the nucleation mode grow to Aitken and accumulation mode, and remain in the atmosphere forming the background aerosol concentration. This makes it impossible to completely remove the effect of new particle formation events. These new analyses are mentioned in the revised manuscript. (3.3 Temporal variation)

The air mass arriving path was also studied as function of the time of day. The only clear pattern was that air masses coming from western sector (most common) were more often coming from high altitude during night than during day. That, however, should not be interpreted as boundary layer / free troposphere pattern, as boundary layer evolution is not included in the trajectory model.

The distribution types are jus a more simple way of presenting the size distribution.

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Therefore they do (and should) contain all the changes in seasonal or diurnal pattern.

The driving factors of the diurnal and seasonal variation are the new particle formation events and the seasonal air mass pattern. These are presented more clearly in the results and conclusions of the revised manuscript.

REFEREE: Specific comments

(p.2055, line 20-25) Size distribution categories are not well defined and it is not clear on which basis they can be considered different. How sure are we that 5 (and not 4 or 6) distributions are needed to describe all cases? Please provide criteria used for differentiating one to the other. It would be certainly interesting to better link the different distributions to environmental conditions (distribution 1: nucleation events, distribution 2: etc…). In particular, is there any distribution connected to high altitude versus low-altitude sorting of air masses mentioned p.2056?

AUTHORS: The separation to different distribution types was made from the whole size distribution with 28 size bins, not only from the three size fraction given in Figure 2. The text was rearranged to avoid misunderstanding in this matter. Different numbers of distribution types from 3 to 7 were tested, but when there were less than 5 distribution types, information was clearly lost. When the number was more than 5, some of the distribution types became very similar, and also the number of cases in some distribution types became quite low.

Distribution types 2 and 5 were associated more with high altitude air, whereas distribution type 4 was observed clearly more often in low altitude air. This was also added to the revised manuscript. (3.4 Trajectory analysis)

REFEREE: P. 2056, line 11. Please check if sorting according to seasons is not a Europeancentred view of Central China climate. Are we sure that this seasonality applies there?

AUTHORS: There is a clear seasonal climate pattern at Mount Waliguan. At summer

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the temperatures are around +10 C and the prevailing winds are from the east and north-east. The low pressure center of the South-Asian monsoon is located above Himalayas and Tibet, south-west from Mount Waliguan. At winter the low pressure is located further south and dry western winds dominate. Winter temperature is around -10 C. Spring and fall are transition periods between these two extremes. Therefore we find it justified to use the four-season sorting to the Mount Waliguan data.

REFEREE: In addition, I am concerned that summer may not be well represented given instrument failures. This issue should be addressed somewhere in the text.

AUTHORS: This issue is addressed in the revised manuscript as a reminder to the reader whenever the summer conditions are brought out.

REFEREE: In table 2 and in most reported averages throughout the text, mean values are reported without corresponding standard deviation. This should be added also for statistically comparing different conditions.

AUTHORS: The average or median values do not tell about variability. However, the standard deviation is not a good parameter to use when describing a tilted distribution such as the occurance of different particle number concentrations (which can not be below zero). Instead of standard deviation the 10 and 90 percentiles are added to most reported values in the revised manuscript.

In Table 2 this was not possible, since the values do not represent the ratios between the concentrations during consecutive nay and night, but the seasonal daytime average (or median) value divided by the nighttime average (or median) value. This was made more clear in the table.

REFEREE: P.2056, line 18-25: Using the CPC instead of the DMPS certainly fills the gap but also bring additional uncertainty (see Fig 3). It is surprising to find CPC values lower than those of DMPS. Excluding new particle formation events from the statistical analysis may then significantly reduce the Ncpc/Ndmps variability. The use of the CPC

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data is not clearly mentioned in the text or in the tables.

AUTHORS: The Ncpc/Ndmps variability is analyzed in the revised manuscript with and without the new particle formation events. The new particle formation periods were defined as periods with Nnuc/Ntot $>$ 0.392 (average + 1 standard deviation of Nnuc/Ntot). The variability was reduced, but still some variability remained. The CPC data is used to check the credibility of the DMPS measurements and in the investigation of the annual variation, but not elsewhere. (3.1 General)

REFEREE: As mentioned in the text, new particle formation is most likely affecting size distribution. This is a very local and intermittent phenomenon. To which extent is this impacting on the mean values of Table 2? Wouldn’t the correct way be removal of all new particle formation events before data processing or to treat them separately? It is surprising to see P.2064, line 10-15 that occurrence of particle nucleation events is derived from correlation between air mass origin and average number of particles in the nucleation mode: analysis of raw DMPS information can be used directly identify nucleation events. Check Venzac et al. (2008, High Frequency New Particle Formation in the Himalayas; PNAS, vol. 105, no. 41, 15666-15671) for additional comparison at high-altitude Asian sites.

AUTHORS: More details (including the reference above) concerning new particle formation events were added to the revised manuscript. New particle formation events at Mount Waliguan were seen directly in the DMPS data, but the detailed analysis of those was not included in this paper. The focus of this paper lies more in the general characteristics of the aerosols in the area. (3.3 Temporal Variation)

New particle formation events last for hours or maximum days, but they happen in large areas (Kulmala et al., 1998, Tellus 50B: 449-462). These events are a source of nucleation mode particles, which then grow to Aitken and accumulation mode sizes. These particles form the stable continental background aerosol level if there are no other particle sources available. Therefore it is impossible to take out the effect of the new

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particle formation events in the aerosol concentrations in an area where they occur. The highest peaks in the number concentration do affect the average concentrations, but not the median values. That is one reason why both average and median values are reported in the manuscript.

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