

**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

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# Mixing of dust and NH<sub>3</sub> observed globally over anthropogenic dust sources

**P. Ginoux<sup>1</sup>, L. Clarisse<sup>2</sup>, C. Clerbaux<sup>2,3</sup>, P.-F. Coheur<sup>2</sup>, O. Dubovik<sup>4</sup>, N. C. Hsu<sup>5</sup>, and M. Van Damme<sup>2</sup>**

<sup>1</sup>NOAA Geophysical Fluid Dynamics Laboratory, Princeton, New Jersey, USA

<sup>2</sup>Spectroscopie de l'Atmosphère, Service de Chimie Quantique et Photophysique, Université Libre de Bruxelles, Brussels, Belgium

<sup>3</sup>UPMC Université Paris 06; Université Versailles St.-Quentin; CNRS/INSU, LATMOS-IPSL, Paris, France

<sup>4</sup>Laboratoire d'Optique Atmosphérique, Université de Lille 1/CNRS, Villeneuve d'Ascq, France

<sup>5</sup>NASA Goddard Space Flight Center, Greenbelt, Maryland, USA

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Correspondence to: P. Ginoux (paul.ginoux@noaa.gov)

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

The global distribution of dust column burden derived from MODIS Deep Blue aerosol products is compared to  $\text{NH}_3$  column burden retrieved from IASI infrared spectra. We found similarities in their spatial distributions, in particular their hot spots are often collocated over croplands and to a lesser extent pastures. Globally, we found 22 % of dust burden collocated with  $\text{NH}_3$ . This confirms the importance of anthropogenic dust from agriculture. Regionally, the Indian subcontinent has the highest amount of dust mixed with  $\text{NH}_3$  (26 %), mostly over cropland and during the pre-monsoon season. North Africa represents 50 % of total dust burden but accounts for only 4 % of mixed dust, which is found over croplands and pastures in Sahel and the coastal region of the Mediterranean. In order to evaluate the radiative effect of this mixing on dust optical properties, we derive the mass extinction efficiency for various mixtures of dust and  $\text{NH}_3$ , using AERONET sunphotometers data. We found that for dusty days the coarse mode mass extinction efficiency decreases from 0.62 to  $0.48 \text{ m}^2 \text{ g}^{-1}$  as  $\text{NH}_3$  burden increases from 0 to  $40 \text{ mg m}^{-2}$ . The fine mode extinction efficiency, ranging from 4 to  $16 \text{ m}^2 \text{ g}^{-1}$ , does not appear to depend on  $\text{NH}_3$  concentration or relative humidity but rather on mineralogical composition and mixing with other aerosols. Our results imply that a significant amount of dust is already mixed with ammonium salt before its long range transport. This in turn will affect dust lifetime, and its interactions with radiation and cloud properties.

## 1 Introduction

Mineral dust affects climate by absorbing and scattering solar and terrestrial radiation, by modifying cloud properties, and by interacting with ozone chemistry and the biosphere (Forster et al., 2007). The amplitude of these effects depends on the size, optical and chemical properties of dust. These properties may be considerably modified when dust is mixed with other aerosol type. In the case of dust mixed with ammonium

## Mixing of dust and $\text{NH}_3$

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



salts, (1) the amount of solar absorption will decrease (Bauer et al., 2007), (2) the activation of cloud condensation nuclei will increase (Levin et al., 1996), (3) the activation of ice nucleation sites will decrease (Sullivan et al., 2010), (4) the recycling of NO<sub>x</sub> is suppressed reducing pollution influence on surface ozone (Fairlie et al., 2010), and (4) iron dissolution which enhances ocean fertilization will increase (Meskhidze et al., 2003). Therefore, it is crucial to determine the mixing state of dust in order to better understand its effects on climate.

In situ observations have shown that ammonium salts commonly accumulate on dust particles (Galy-Lacaux et al., 2001; Jordan et al., 2003; Sullivan et al., 2007; Shi et al., 2008). The coating of dust particle by sulfate occurred by in-cloud processes, by which reactions with sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) initiate the deposition of sulfate on the surface of the particles (Levin et al., 1996). Other mixing processes include coagulation of sulfate particles or oxidation of sulfur dioxide (SO<sub>2</sub>) to sulfate on the particle surface in the presence of ammonia (NH<sub>3</sub>). The latter process involves titration of NH<sub>3</sub> by H<sub>2</sub>SO<sub>4</sub> to form ammonium sulfate (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or bisulfate (NH<sub>4</sub>)SO<sub>4</sub>. Similarly, in presence of nitric acid (HNO<sub>3</sub>), NH<sub>3</sub> will produce ammonium nitrate (NH<sub>3</sub>NO<sub>3</sub>) (Usher et al., 2003). Sulfate coatings on dust undergo efflorescence (crystallization) at different relative humidities depending on the size and mineralogy of dust (Usher et al., 2003).

By analyzing Asian dust, Sullivan et al. (2007) showed that transport pathways of dust through source of pollutants is more important than reaction kinetics in determining which species accumulate on dust. The scarcity of in-situ observations makes it difficult to extrapolate the predominance of dust mixing with pollutants at global scale, such that the impact of such mixing on climate is evaluated using poorly constrained models (Lesins et al., 2002; Bauer et al., 2007).

Using the Moderate Resolution Imaging Spectroradiometer (MODIS) Deep Blue satellite products, Ginoux et al. (2010) have retrieved global dust sources at 0.1° resolution. By comparing their location with land-use fraction, Ginoux et al. (2012) have calculated that 25% of dust is emitted from agriculture, and called such dust as *anthropogenic*. Meanwhile, Clarisse et al. (2009) have retrieved the global distribution of

ammonia (NH<sub>3</sub>) from the Infrared Atmospheric Sounding Interferometer (IASI) infrared spectra and have shown that hot spots are often associated with agriculture. There is a remarkable similarity in the observed distribution of these hot spots and anthropogenic dust sources, which may provide the first evidence of co-located pollutants and dust over their sources (see Sect. 3). This would first confirm the existence of anthropogenic dust (Sect. 4), once our results have been validated (Sect. 5), and second that anthropogenic dust has different chemical and optical properties (Sect. 6). The implications of these results on our understanding of the effects of dust on climate are discussed in Sect. 7.

## 2 Satellite data

### 2.1 Dust column burden

The MODIS instrument on the polar orbiting Aqua satellite measures daily backscattering radiances globally since 2002. The MODIS Deep Blue algorithm employs radiances from the blue channels of MODIS instruments. At these wavelengths the surface reflectance is very low so that the presence of aerosol is detected by an increase of total reflectance and enhanced spectral contrast (Hsu et al., 2004, 2006). The values of AOD and single scattering albedo ( $\omega$ ) at 412, 470, 550 and 670 nm, and the Angstrom exponent ( $\alpha$ ) between 412 and 470 nm are retrieved at the pixel level over bright surfaces (reflectance at 550 nm greater than 0.15), and then averaged on a 10 by 10 km grid. The data are aggregated into granules forming the Level 2 data. These data are interpolated by surface-weighted averaging on a 0.1° by 0.1° regular latitude/longitude grid. Ginoux et al. (2012) extracted Dust Optical Depth (DOD) from MODIS collection 5.1 Level 2 (M-DB2) aerosol products by requiring the values of the Angstrom exponent to be less than 0, and the spectral variation of the single scattering albedo to increase with wavelength. By analyzing 7 yr of daily global DOD, Ginoux et al. (2012) also found that globally 24 % of dust is emitted from sources associated with land-use fraction

## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



greater than 30 %, which they defined as anthropogenic. They also found very large variations between continents and seasons.

In the present study, we use daily global M-DB2 DOD from April 2009 to March 2010 and convert it into column burden. The conversion is performed following Ginoux et al. (2001) who expressed DOD ( $\tau$ ) and mass burden  $M$  by the following relation,

$$M = \frac{4 \rho r_{\text{eff}}}{3 Q_{\text{ext}}} \tau = \frac{1}{\epsilon} \tau \quad (1)$$

where  $r_{\text{eff}} = 1.2 \times 10^{-6}$  m is the effective radius,  $\rho = 2600 \text{ kg m}^{-3}$  is the density of dust,  $Q_{\text{ext}} = 2.5$  is the extinction efficiency at 550 nm and for a  $1.2 \times 10^{-6}$  particle radius,  $\epsilon = 0.6 \text{ m}^2 \text{ g}^{-1}$  is the mass extinction efficiency, and  $\tau$  is dust optical depth at 550 nm. The selection of  $r_{\text{eff}}$  is based on the effective radius of the total size distribution measured at Sal Island (Cape Verde) and Banizoumbou (Niger) by Tanré et al. (2001). As the aerosol optical depth increase, they observed that  $r_{\text{eff}}$  reaches  $1.2 \times 10^{-6}$  m at these two sites.

Such relation was also used by Kaufman et al. (2005) to convert DOD to  $M$ . Using  $r_{\text{eff}} = 1.7 \times 10^{-6}$  m,  $\rho = 2600 \text{ kg m}^{-3}$  and  $Q_{\text{ext}} = 2.2$ , they obtained  $\epsilon = 0.37 \pm 0.05 \text{ m}^2 \text{ g}^{-1}$ , which is 40 % lower than our value. On the other hand, our value is closer to the median value ( $0.72 \text{ m}^2 \text{ g}^{-1}$ ) derived from global dust models (Huneus et al., 2011). The range of values among these models is quite large, varying from 0.25 to  $1.28 \text{ m}^2 \text{ g}^{-1}$ , which is indicative of large uncertainty in  $\epsilon$ . Similar range has been obtained from theoretical calculations (Hansell et al., 2011), with higher values associated with non-spherical particles. Also, values derived from field campaigns vary from  $0.6 \pm 0.1$  at Barbados (Li et al., 1996) to  $1.09 \pm 0.4$  at Cape Verde (Chen et al., 2011). There is clearly a need to better constrain  $\epsilon$ , particularly over source areas where there are fewer data. This will be discussed in Sect. 5.

**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 2.2 NH<sub>3</sub> column burden

Infrared radiances have been measured daily since the end of 2006 by the IASI instrument on board the EUMETSAT's polar orbiter MetOp. Clarisse et al. (2009) calculated the brightness temperature difference between radiances and converted it to column burden of NH<sub>3</sub>. The first global distributions were obtained by averaging one year of measurements and 28 hotspot regions were identified. Here we use an improved product only available from April 2009 to March 2010. For this product NH<sub>3</sub> column burden is retrieved using a full Optimal Estimation scheme, as in Clarisse et al. (2010a), and similar to the processing algorithm used for other trace gases (Hurtmans et al., 2012). The retrieval is only carried out for the observations with a detectable spectral signal of NH<sub>3</sub>. Hence, very low concentrations will not be measured. Similarly, when the contrast in temperatures between the ground and the atmosphere is weak, the IASI sensitivity to surface concentrations is close to zero, and even significant NH<sub>3</sub> concentrations might be missed. While the IASI optimal estimation retrievals account for emissivity features by using temporal and spectrally resolved emissivity data (Zhou et al., 2011), some residual contamination due to emissivity features of windblown dust (Clarisse et al., 2010a) cannot be excluded. To minimize these, only those NH<sub>3</sub> retrievals with root mean square of the observed minus fitted spectrum below  $2.8 \text{ W cm}^{-1} \text{ sr}^{-1}$  were retained. This threshold value is close to the IASI instrumental noise in the relevant spectral range (Clerbaux et al., 2009).

Here we use the daily NH<sub>3</sub> column burden retrieved from April 2009 to March 2010 on each pixel (12 km diameter at nadir). Monthly averages are computed on a  $0.25^\circ$  by  $0.25^\circ$  grid. The value in each grid cell was obtained by averaging all observations within that cell. In grid cells where no measurements were made a value of zero was assigned. Although some biases cannot be avoided, this approach minimizes artifacts that would be introduced from interpolation of sparse data.

### Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



### 3 Global distribution of anthropogenic dust and NH<sub>3</sub>

Using Klein Goldewijk (2001) land-use database, we distinguish between natural and anthropogenic (agricultural) dust by assuming that the latter corresponds to dust over areas with more than 30 % land-use. Natural dust is associated with less than 30 % land-use. We further separate between cropland and pasture dust depending on which one has the largest fraction within a grid cell.

Figure 1 shows the annual mean column burden of M-DB2 dust, and IASI NH<sub>3</sub>. We see that most of the “dust belt”, defined by Prospero et al. (2002), is covered with at least 0.1 gm<sup>-2</sup> of dust. The Sahara desert is essentially characterized by natural dust while most of Sahel is associated with pasture except in Senegal, Burkina Faso, Niger and Nigeria where cropland dominates. In the Middle East, cropland is confined to some areas of Mesopotamia. In Central Asia, most anthropogenic dust is associated with pasture, while dust over cropland covers most of India and East China. In the Southern Hemisphere, dust over cropland is found in Argentina and Southeast Australia, and dust over pasture covers Patagonia and Central and Northeast Australia. Over the United States, dust over cropland is confined to the southern part of the San Joaquin Valley of California and the Midwest, and dust over pasture dominates the Southwest.

Inventory of global ammonia emissions indicates that 60 % comes from agriculture, with 1/3 from fertilizer and 2/3 from livestock sources (Beusen et al., 2008). Lifetime of gaseous NH<sub>3</sub> is about 22 h (Adams et al., 1999), which produces well defined hot spots centered over its sources. This is clearly apparent in Fig. 1 where NH<sub>3</sub> distribution is covering most cropland, which happens to be also source of dust (i.e. US Great Plains, Argentina, Sahel, Mediterranean coast, Mesopotamia, Uzbekistan, India, East China). The most pronounced feature is over the Indo-Gangetic basin, where dust and NH<sub>3</sub> column burden are greater than 0.5 gm<sup>-2</sup>, and 0.5 mgm<sup>-2</sup>, respectively.

The global annual mean dust burden from M-DB2 is 4.2 Tg, which is four times lower than the median value of 16 Tg derived by Huneus et al. (2011) from 14 models,

## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



with values ranging from 6.8 to 29.5 Tg depending on the model. There are two major reasons for this discrepancy: one is due to uncertainty in  $\epsilon$  in Eq. (1) and the other comes from M-DB2 retrieval over bright surface only. By excluding dark surfaces (i.e. oceans) transported dust is not included in our results. As we are interested to study dust over its sources, which are located over land, this is not an issue. Additionally, we consider only one year while interannual variability of dust burden is of the order of 20 % (Ginoux et al., 2004).

#### 4 Anthropogenic dust mixed with NH<sub>3</sub>

In this section, we quantify the mixing of dust with NH<sub>3</sub> over agriculture at the global and continental scales.

The percentage of anthropogenic dust mixed with NH<sub>3</sub> over each landcover types is calculated by summing areas of each type where M-DB2 dust is mixed with detectable amount of IASI NH<sub>3</sub>, and dividing by the total surface of that cover type where M-DB2 dust is present. We see in Fig. 2 that the largest percentage of mixed dust with NH<sub>3</sub> is over cropland globally, and for each sub-continent.

Table 1 provides more details on the annual burden and fraction of dust mixed with NH<sub>3</sub> over various continental regions. Globally, 7 % of dust is mixed with NH<sub>3</sub>, which represents 310 ktons of dust. If we consider only croplands, we found 22 % of dust is mixed with NH<sub>3</sub>, which corresponds to 115 ktons or 3 % of global dust burden (4.2 Tg). The largest fraction of dust mixed with NH<sub>3</sub> (26 %) is observed over South and Central Asia subcontinent, with 34 % over cropland, 5 % over pasture, and 12 % over natural sources. The other area with significant amount of mixed dust is East Asia (67 ktons or 14 % of global burden) with 25 % from cropland. In North America, mixed dust represents only 14 ktons (11 % of North American dust burden) mostly from cropland (19 %). North Africa represents 46 % of global dust burden, but only 77 ktons (less than 2 % of global burden) is mixed with NH<sub>3</sub> with most of it over cropland.

## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



## 5 Comparison with AERONET data

In order to evaluate M-DB2 derived column burden of dust, its value is compared with retrieval from inversion of Aerosol Robotic Network (AERONET) sunphotometer data. AERONET is a federated worldwide network of sunphotometers that are monitored and maintained at the NASA Goddard Space Flight Center (Holben et al., 1998). Dust particles are essentially greater than  $1\ \mu\text{m}$  radius (coarse mode), but about 10% of dust mass, mostly clay, is submicron (fine mode) (Ginoux et al., 2001). The parameters of the size distribution for fine and coarse modes are retrieved by inversion of Almuantar measurements (Dubovik and King, 2000). Here we use the daily quality assured (Level 2) volume concentration for the coarse mode from 1 April 2009 to the end of March 2010 in order to coincide with IASI data.

The Angstrom wavelength exponent  $\alpha$  is often used as an indicator of the dominance of one aerosol mode over the other. For example, using AERONET data, Eck et al. (1999) have shown that  $\alpha$  values are generally lower than 0.5 in dusty environments and higher in polluted regions. To make sure that dust is the major aerosol contributing to the measured extinction, we extract coarse mode volume concentration when  $\alpha < 0.5$ . The volume concentration is then multiplied by the dust density ( $2600\ \text{kg m}^{-3}$ ) to convert it into column burden.

Ten AERONET sites have been selected based on the diversity of their geographical location over agriculture dominated areas. Satellite data within 25 km of the AERONET sites are used for this comparison.

Figure 3 shows the time series of daily and monthly dust column burden from M-DB2 and AERONET and monthly IASI  $\text{NH}_3$  column burden at the selected sites from April 2009 to March 2010. The largest root mean square difference (RMSD) between AERONET and M-DB2 dust burden appears in Niger at Banizoumbou (RMSD =  $0.44\ \text{g m}^{-2}$ ) and Zinder (RMSD =  $0.41\ \text{g m}^{-2}$ ), but the mean values are also the largest at these sites. The highest monthly dust burden ( $2.3\ \text{g m}^{-2}$ ) among all ten sites is observed in Banizoumbou (Niger) in March. African sites are characterized by

### Mixing of dust and $\text{NH}_3$

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



two maxima of dust burden, one in March and the other in boreal summer (June-July-August). The seasonal variation of dust burden at the sites located in Sahel (south of 20° N) can be explained by the movement of the InterTropical Convergence Zone (ITCZ) occupying its southernmost position (5° N) in December and its northernmost position (20° N) in July. As the ITCZ moves over these sites, dry air mass from the north is replaced by monsoon flow from the south bringing precipitation, which suppress dust emission and removes dust particles from the atmosphere. Sharp changes of wind direction and relative humidity have been observed in mid-October and early May at Banizoumbou, as well as a maximum of dust optical depth in March and June (Rajot et al., 2008). Summer maximum of dust load at the Kuwait site has been associated with increased surface winds in summer (Sabbah, 2010). The RMSD (0.22 gm<sup>-2</sup>) is relatively low at site, but is reaching 0.3 gm<sup>-2</sup> in Sede Boker (Israel). The summer maximum in Sede Boker has been associated with dust originating from the Arabian Peninsula and the Sahara (Israelevich et al., 2003). At the two sites in the Indo-Gangetic basin (Karachi and Jaipur), there is a pronounced maximum of dust load during the pre-monsoon season with monthly dust burden reaching up to 1.5 gm<sup>-2</sup>. This seasonal variation corresponds to the climatological (2001–2009) cycle of coarse mode optical depth measured with sunphotometer over the Indo-Gangetic Plain (Giles et al., 2011). In China, dust activity peaks in spring (Prospero et al., 2002) but M-DB2 value underestimates AERONET data by a factor 2 in March. This large discrepancy may be due to the low number of observations during that month.

The summer maximum of IASI NH<sub>3</sub> observed in Maricopa has been attributed to the strong dependency of ammonia volatilization on surface temperature (Clarisse et al., 2010b). The similarity of NH<sub>3</sub> and dust cycle suggests also a dependency on wind speed on NH<sub>3</sub> volatilization.

In Africa, there is a distinct peak of IASI NH<sub>3</sub> in May which was also observed in surface concentration of NH<sub>3</sub> in Banizoumbou by Galy-Lacaux et al. (2001). They related the high concentration of ammonia in Banizoumbou to a local density gradient of animals grazing in pasture areas, but they did not discuss the origin of this pronounced

5 peak in May. Except for this peak, the seasonal cycle of  $\text{NH}_3$  in Africa is quite similar to the one of dust. The highest concentration of  $\text{NH}_3$  is observed in India. The high level of  $\text{NH}_3$  concentration in India compare to the rest of the world has already been shown by Dentener and Crutzen (1994) and later confirmed by Clarisse et al. (2009). The similarity of dust and  $\text{NH}_3$  seasonal cycles at all these different sites, except for Karachi, suggests a common origin, such that  $\text{NH}_3$  may be used as a tracer for anthropogenic dust. Next, we would like to detect potential difference in optical properties between natural and anthropogenic dust, in particular concerning the mass extinction efficiency.

## 6 Mass extinction efficiency of dust

### 10 6.1 Dependency on $\text{NH}_3$

Theoretical calculation using Mie theory indicates that the mass extinction efficiency  $\epsilon$  of dust decreases when mixed with sulfate or nitrate (Bauer et al., 2007). As we have seen that  $\text{NH}_3$  is a good tracer for anthropogenic dust, the influence of anthropogenic dust on  $\epsilon$  should be correlated with  $\text{NH}_3$  concentration.

15 To examine the existence of such effect on optical properties retrieved from sunphotometers, we extract all AERONET inversion Level 2 daily data from April 2009 to March 2010. As we are only interested in dusty cases, we restrict the data for days when  $\alpha$  is lower than 0.5. To calculate the mass extinction efficiency we multiply the volume concentration retrieved from the inversion of AERONET data (Dubovik and King, 2000) by the mass density of dust ( $2600 \text{ kg m}^{-3}$ ) and divide by the extinction measured directly by the sunphotometer. We selected the extinction at 670 nm because there are more instruments measuring at that wavelength than at 550 nm. The fine and coarse mode mass extinction efficiencies are calculated separately, using the corresponding retrieved volume concentration. These values are then clustered as a function of co-located (same day and within 25 km) IASI  $\text{NH}_3$  concentration.

20

25

## Mixing of dust and $\text{NH}_3$

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Figure 4 shows the daily values of mass extinction efficiency  $\epsilon$  of fine (FMEE) and coarse (CMEE) modes at all AERONET sites with Angstrom exponent less than 0.5, as a function of NH<sub>3</sub> column burden and season. The mean values of FMEE and CMEE for different ranges of NH<sub>3</sub> concentrations are also provided in the Figure. We see a clear decrease of CMEE as NH<sub>3</sub> increases, with mean values at 0.62 m<sup>2</sup>g<sup>-1</sup> for NH<sub>3</sub> less than 2 mgm<sup>-2</sup> and decreasing to 0.48 m<sup>2</sup>g<sup>-1</sup> for NH<sub>3</sub> greater than 20 mgm<sup>-2</sup>. On the other hand, there is a factor 3 variability of FMEE but apparently unrelated to NH<sub>3</sub> concentration. Considering NH<sub>3</sub> as a tracer of anthropogenic dust, these results suggest that the mass extinction efficiency of anthropogenic dust has a lower coarse mode mass extinction efficiency than natural dust, but has similar fine mode mass extinction as natural dust. So, the value of  $\epsilon = 0.6 \text{ m}^2 \text{ g}^{-1}$  derived from Eq. (1) corresponds quite well to the mean value observed by AERONET sunphotometer in the case of natural dust.

Figure 4 shows that the variability of FMEE at Zinder Airport in summer is as large as a factor 3, still with the same range of NH<sub>3</sub> burden. Similarly, FMEE at Jaipur in spring varies by more than a factor 2 while NH<sub>3</sub> is ranging between 2 and 4 mgm<sup>-2</sup>. There is clearly another factor controlling FMEE variability.

Sullivan et al. (2007) have shown that ammonium sulfate will preferentially accumulate on fine mode dust because H<sub>2</sub>SO<sub>4</sub> reactions with dust depends on aerosol surface area to which fine mode contributes more than coarse mode. They further explained that these reactions will displace any preexisting nitrate back to the gas phase and will subsequently accumulate on the coarse mode. Ammonium salts were found to be internally mixed with dust, and are strongly correlated with the sum of acids in dust. Thus they demonstrated that ammonium predominantly accumulates in fine mode dust, and subsequently on coarse mode with increasing ammonium. As ammonium salts are hygroscopic particles, if they are mixed with dust then FMEE should be sensitive to relative humidity, which may explain the large variation of FMEE at one location and during the same season.

## 6.2 Dependency on RH

Hygroscopic growth of ammonium sulfate can increase  $\epsilon$  by an order of magnitude from its crystallization point to high ( $> 90\%$ ) relative humidity (Tang, 1996). We should also expect some growth of fine dust internally mixed with sulfate, although not as pronounced as pure sulfate because dust itself is hydrophobic.

To verify that fine mode dust is mixed with  $\text{NH}_3$  and grows hygroscopically, we extract the daily boundary layer (925 hPa) relative humidity (RH) from NCEP/NCAR re-analysis (Kalnay et al., 1996) at the AERONET sites for the corresponding days and locations. Figure 5 (upper panel) is similar to Fig. 4 but each data is color coded as a function of RH. To our surprise there is no apparent dependency of mean FMEE on RH although RH varies below and above the deliquescence point of  $(\text{NH}_4)_2\text{SO}_4$  (Haywood and Ramawsamy, 1998). At Zinder airport, the relative humidity in summer does not vary considerably ranging from 85 and 90%. Similarly, at the Jaipur site in spring, RH is below the deliquescence point as RH values varies between 75 and 80%. These results indicate the weak dependency of FMEE on hygroscopic growth, but it does not mean that there is none. Indeed, fine mode aerosols are mostly sensitive to change in scattering and absorption properties rather than size. The opposite is true for the coarse mode (Seinfeld and Pandis, 1998). So, it may be more effective to study variability of FMEE based on absorption property.

## 6.3 Dependency on $k$

Mass extinction efficiency of aerosol depends on the refractive index, whose imaginary part is related to its absorption property. Figure 5 (lower panel) shows the dependency of FMEE and CMEE on the imaginary part of the refractive index ( $k$ ) at 670 nm. As absorption increases FMEE decreases from about 12 to  $5\text{ m}^2\text{ g}^{-1}$ . The decrease of FMEE with increasing  $k$  appears clearly at the two AERONET sites (Zinder airport and Jaipur).

### Mixing of dust and $\text{NH}_3$

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Analysis of dust samples collected at different locations in North Africa indicates  $k$  values at 655 nm ranging from 0.0016  $i$  to 0.0065  $i$  (Wagner et al., 2012), which is covering most of our results. The size resolved composition of the soil samples by Wagner et al. (2012) indicates a much larger abundance of sulfate and other non-crustal elements in the fine mode, with sulfate volume abundance varying from 2 to 6%. The authors related the variability of  $k$  to mineralogical composition. In fact, mineralogy determines not only the amount of iron and thus absorption by dust but also the chemical processing by ammonium salts. Sullivan et al. (2007) have shown that sulfate is more strongly associated with aluminosilicate-rich dust particles while nitrate and chloride are more associated with calcite-rich dust. Iron is essentially associated with aluminosilicates. This double influence of mineralogy may explain the variability of FMEE. The strong dependency of  $k$  to source location has been shown for Sharan dust using backtrajectory analysis (McConnell et al., 2010). Here, we show that such variation is also linked to the anthropogenic origin of dust due to its mixing with ammonium salts or other acidic components from organic soils used for agriculture.

We can conclude from Figs. 4 and 5, that over source areas, natural dust mass extinction efficiency is  $0.6 \text{ m}^2 \text{ g}^{-1}$  for coarse mode, and decreases to  $0.4\text{--}0.5 \text{ m}^2 \text{ g}^{-1}$  for anthropogenic dust, mostly due to mixing with ammonium salts or organics from fertilizer. On the other hand, for fine mode dust mineralogy is a major factor controlling the mass extinction efficiency.

## 7 Conclusions

The global distributions of dust and NH<sub>3</sub> column burden have been compared to determine: (1) if they are mixed over dust sources, (2) the global and regional fraction of dust mixed with NH<sub>3</sub>, and (3) the effect of such mixing on dust optical properties.

To achieve these objectives, dust optical depth (DOD) derived from M-DB2 aerosol products has been converted into column burden by dividing DOD by a theoretical

mass extinction efficiency. The distribution of M-DB2 dust column burden is then compared to  $\text{NH}_3$  retrieved from the IASI satellite data.

Using land-use dataset we found substantial similarity between the annual mean distribution of dust and  $\text{NH}_3$  burdens, particularly over croplands. The mass of dust mixed with  $\text{NH}_3$  represents 7% of the global mass estimated to be 4.1 Tg over dust sources. Over croplands mixed dust represents 22% globally. On a seasonal basis, the maximum mixing is observed in spring with almost 30% of dust over cropland mixed with  $\text{NH}_3$ . Among the different continents, the Indian subcontinent has the largest fraction of dust mixed with  $\text{NH}_3$  (26%). Our results indicate that  $\text{NH}_3$  is a good tracer for anthropogenic dust.

Comparison of dust column burden retrieved from M-DB2 and sunphotometers at 10 AERONET sites, spread over different continents, indicates good agreement in Africa and Asia but large overestimation of M-DB2 values over Arizona and to a lesser extent in Israel.

To study the difference between anthropogenic and natural dust on optical properties, we derived the mass extinction efficiency for the coarse and fine mode at all AERONET sites. Only dusty days with Angstrom exponent less than 0.5 are considered. By collocating IASI  $\text{NH}_3$  column burden with AERONET data, we found that  $\epsilon$  is  $0.6 \text{ m}^2 \text{ g}^{-1}$  for the coarse mode, and it decreases to a minimum of  $0.4\text{--}0.5 \text{ m}^2 \text{ g}^{-1}$  when mixed with concentration of  $\text{NH}_3$  in excess of  $20 \text{ mg m}^{-2}$ . This represents a 20% decrease with a ten-fold increase of  $\text{NH}_3$  concentration. The mass extinction efficiency for fine mode is ten times higher than for coarse mode of unmixed dust, with no clear dependency on  $\text{NH}_3$  concentration or relative humidity. Instead, we found a sensitivity to the imaginary part of the refractive index that we attribute to mineralogy and mixing with other aerosols. In view of previous analysis of in-situ data, we related this decrease of coarse mode  $\epsilon$  as a result of the accumulation of purely scattering ammonium, while for the fine mode the amount of absorption is determined by the mineralogical composition. Mineralogical composition determines not only the amount of absorptive iron but also the accumulation of ammonium salts.

## Mixing of dust and $\text{NH}_3$

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The relatively weak change of mass extinction efficiency is in agreement with a previous model study (Bauer et al., 2007). However, such study considered dust aging during transport whereas we consider here dust co-located mostly over cropland with IASI NH<sub>3</sub> hot spots. The existence of mixed dust with NH<sub>3</sub> over large areas implies that a significant amount of dust is already mixed with ammonium salt before its long range transport and cloud processing. This in turn will affect dust lifetime, and its interactions with radiation, cloud properties, and ocean biogeochemistry.

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## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Tanré, D., Kaufman, Y. J., Holben, B. N., Chatenet, B., Karnieli, A., Lavenu, F., Blarel, L., Dubovik, O., Remer, L. A., and Smirnov, A.: Climatology of dust aerosol size distribution and optical properties derived from remotely sensed data in the solar spectrum, *J. Geophys. Res.*, 106, 18205–18217, doi:10.1029/2000JD900663, 2001. 12507
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**Mixing of dust and  
NH<sub>3</sub>**

P. Ginoux et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## Mixing of dust and NH<sub>3</sub>

P. Ginoux et al.

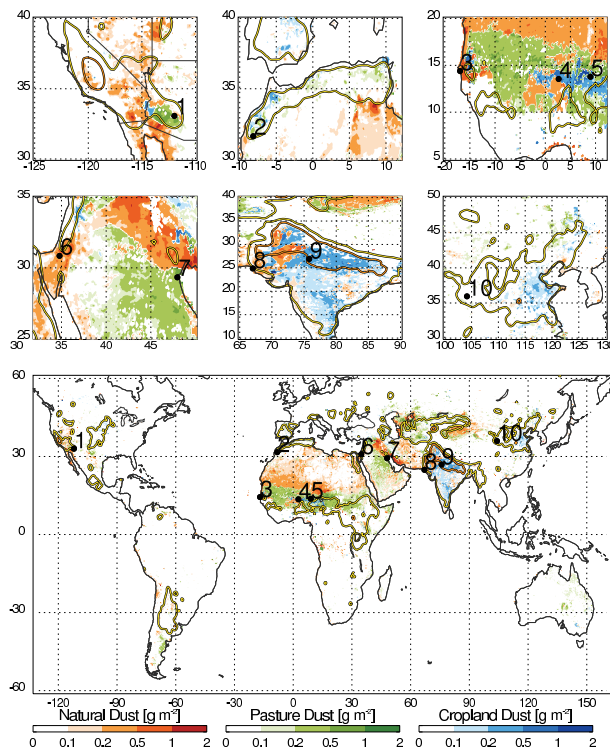
**Table 1.** Annual mean total burden of M-DB2 dust ( $M_{DU}$ ) in units of ktons as well as the fraction (%) of dust mixed with NH<sub>3</sub> over all land cover types ( $f^{tot}$ ), cropland ( $f^{crop}$ ), pasture ( $f^{past}$ ), and with less than 30 % land-use ( $f^{nat}$ ) over 9 regions which boundaries are defined by their minimum and maximum latitude (Lat) and longitude (Lon). Percentage fraction greater than 20 % are indicated in bold font.

Region	Lon Range	Lat Range	$M_{DU}^{tot}$	$M_{DU}^{crop}$	$M_{DU}^{past}$	$M_{DU}^{nat}$	$f^{tot}$	$f^{crop}$	$f^{past}$	$f^{nat}$
Global	130° W–160° E	60° S–60° N	4184	532	1622	2029	7	<b>22</b>	7	4
North America	125° W–70° W	20° N–50° N	128	9	85	33	11	19	11	10
South America	85° W–60° W	55° S–0° N	70	3	28	38	6	<b>26</b>	3	6
North Africa	20° W–35° E	5° N–40° N	1934	141	667	1126	4	11	7	2
South Africa	5° E–50° E	35° S–5° N	95	2	70	24	5	13	5	4
West Asia	35° E–60° E	5° S–50° N	1000	72	431	498	4	8	5	4
Central Asia	60° E–100° E	5° N–30° N	334	223	0	111	<b>26</b>	<b>34</b>	5	12
East Asia	60° E–140° E	30° N–50° N	481	63	222	196	14	<b>25</b>	15	9
Australia	110° E–155° E	45° S–10° S	142	6	133	3	1	3	1	0

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[I◀](#)
[▶I](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


## Mixing of dust and $\text{NH}_3$

P. Ginoux et al.

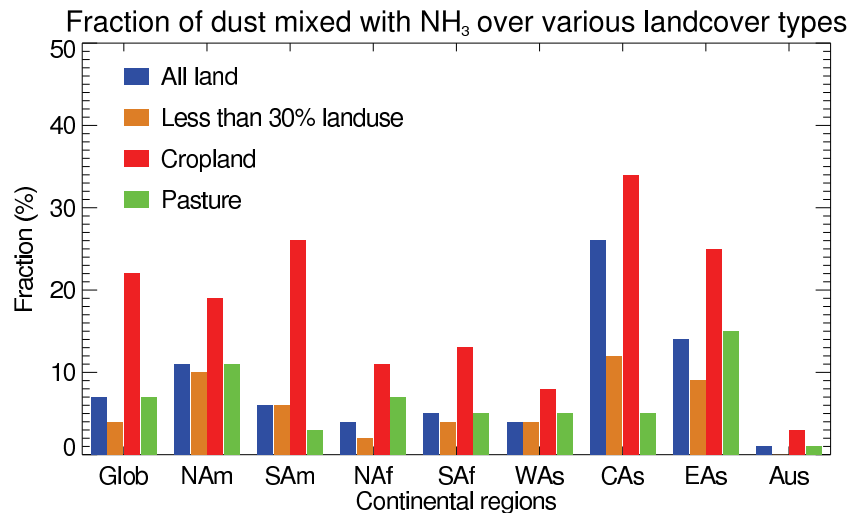


**Fig. 1.** Global (lower panel) annual mean distribution of M-DB2 dust column burden ( $\text{g m}^{-2}$ ) over areas with less than 30% (Natural Dust; red shading) and more than 30% land-use -with an additional distinction between the prevalence of pasture over cropland (pasture dust; green shading) and cropland over pasture (cropland dust; blue shading), 0.5 (yellow contour) and 2.5 (orange contour) column burden ( $\text{mg m}^{-2}$ ) of IASI  $\text{NH}_3$ , with the numbers 1 to 10 indicating the position of the AERONET sites of Fig. 3, and the upper panels providing a zoom over 6 regions.



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P. Ginoux et al.



**Fig. 2.** Annual mean fraction (%) of M-DB2 dust column burden mixed with IASI  $\text{NH}_3$  over all surfaces (blue), areas with less than 30 % land-use (brown), cropland (red), and pasture (green) globally and for 8 continental regions.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

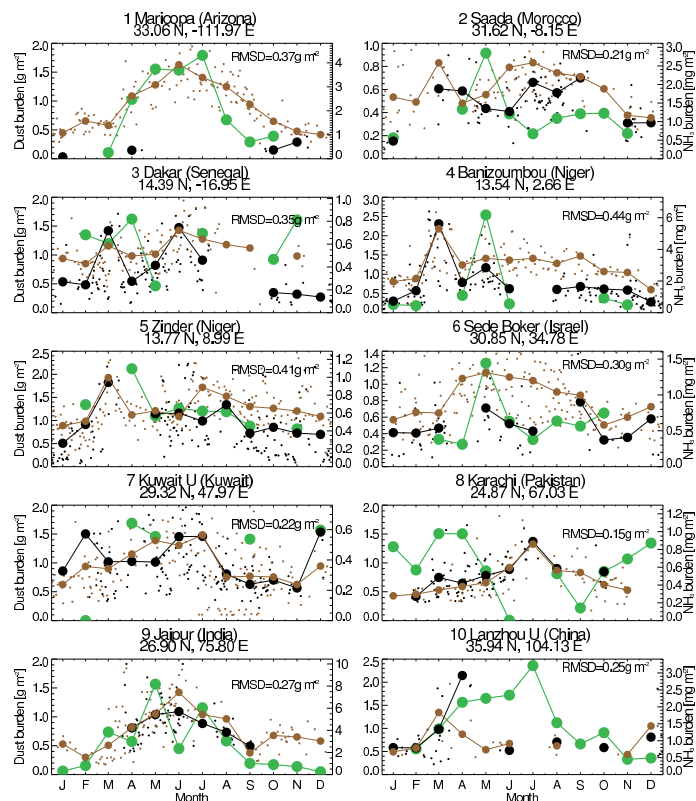
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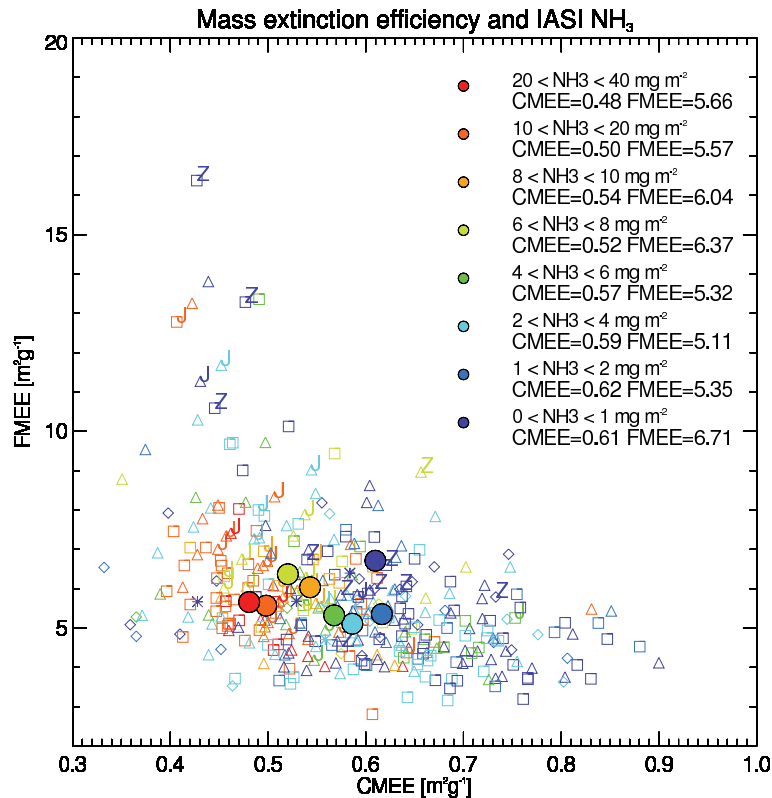
## Mixing of dust and $\text{NH}_3$

P. Ginoux et al.



**Fig. 3.** Daily (small dots) and monthly (large dots) AERONET coarse mode column burden (black), M-DB2 dust burden (brown) in units of  $\text{g m}^{-2}$  (left vertical axes), and IASI  $\text{NH}_3$  column burden (green) in units of  $\text{mg m}^{-2}$  (right vertical axes) at 10 AERONET sites. The root mean square difference (RMSD) in units of  $\text{g m}^{-2}$  between monthly M-DB2 and AERONET mass column burden is provided in the upper right.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

**Fig. 4.** Daily mass extinction efficiency ( $\text{m}^2 \text{g}^{-1}$ ) for coarse (CMEE) and fine (FMEE) modes at all AERONET stations from April 2009 to March 2010 when the Angstrom exponent is less than 0.5, and as a function of IASI  $\text{NH}_3$  column burden for 8 ranges of values (colors), with mean values of FMEE and CMEE given in units of  $\text{m}^2 \text{g}^{-1}$  provided in upper right. The corresponding season is indicated by star (December-January-February), triangle (March-April-May), square (June-July-August), and diamond (September-October-November) symbols. The Zinder Airport and Jaipur AERONET sites are indicated by the letter Z and J, respectively.

Mixing of dust and  $\text{NH}_3$

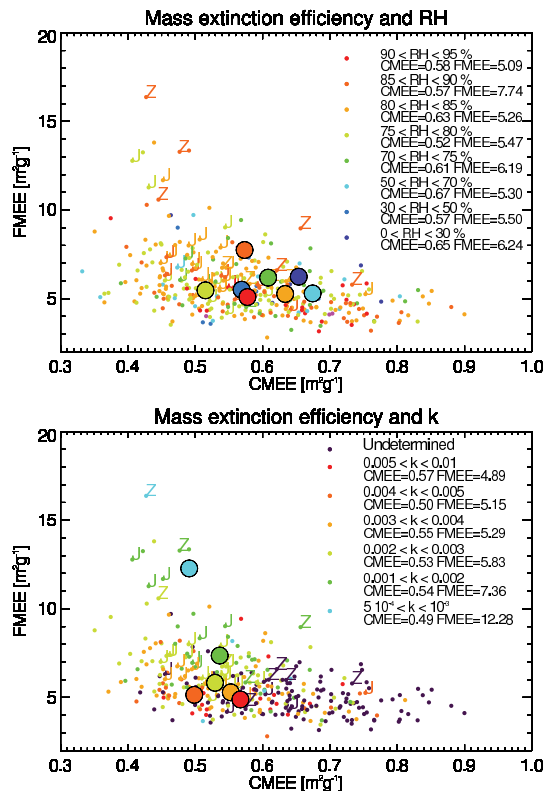
P. Ginoux et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



## Mixing of dust and $\text{NH}_3$

P. Ginoux et al.



**Fig. 5.** Daily mass extinction efficiency ( $\text{m}^2 \text{g}^{-1}$ ) for coarse (CMEE) and fine (FMEE) modes at all AERONET stations from April 2009 to March 2010 when the Angstrom exponent is less than 0.5, and as a function of relative humidity at 925 mb (upper panel) and AERONET imaginary part of refractive index at 670 nm (lower panel). The range of RH and k values and the corresponding mean values of FMEE and CMEE, in units of  $\text{m}^2 \text{g}^{-1}$  are provided on the upper right of each panels. The Zinder Airport and Jaipur AERONET sites are indicated by the letter Z and J, respectively.