

The first estimates of global nucleation mode aerosol concentrations based on satellite measurements

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Received: 26 May 2011 – Published in Atmos. Chem. Phys. Discuss.: 1 July 2011

Revised: 29 September 2011 – Accepted: 22 October 2011 – Published: 2 November 2011

Abstract. Atmospheric aerosols play a key role in the Earth's climate system by scattering and absorbing solar radiation and by acting as cloud condensation nuclei. Satellites are increasingly used to obtain information on properties of aerosol particles with a diameter larger than about 100 nm. However, new aerosol particles formed by nucleation are initially much smaller and grow into the optically active size range on time scales of many hours. In this paper we derive proxies, based on process understanding and ground-based observations, to determine the concentrations of these new particles and their spatial distribution using satellite data. The results are applied to provide seasonal variation of nucleation mode concentration. The proxies describe the concentration of nucleation mode particles over continents. The source rates are related to both regional nucleation and nucleation associated with more restricted sources. The global pattern of nucleation mode particle number concentration predicted by satellite data using our proxies is compared qualitatively against both observations and global model simulations.

1 Introduction

Atmospheric aerosol particles affect the quality of our life in many different ways. First of all, they influence the Earth's radiation balance directly by scattering and absorbing solar radiation, and indirectly by acting as cloud condensation nuclei (e.g. Forster et al., 2007; Myhre, 2009; Quaas

et al., 2009). Secondly, aerosol particles modify the intensity and properties of radiation reaching the Earth's surface, having direct influences on the vegetation and its interactions with the carbon cycle and atmospheric chemistry (Gu et al., 2002; Wang et al., 2008). Thirdly, aerosol particles deteriorate human health and reduce visibility in urban areas (Pope and Dockery, 2006; Hand and Malm, 2007; Anderson, 2009). The various effects of atmospheric aerosol particles are tightly connected via physical, chemical, meteorological and biological processes occurring in the atmosphere and at the atmosphere-biosphere interface (e.g. Arneth et al., 2010). In addition these natural processes and feedbacks, the couplings between atmospheric aerosol particles, trace gases, air quality and climate are affected by human actions, such as emission policy, forest management and land use change (Brasseur and Roeckner, 2005; Arneth et al., 2009; Jacob and Winner, 2009; Raes et al., 2010).

Quantifying the climatic, health and other effects of atmospheric aerosol particles requires detailed information on their physical and chemical properties as well as on their spatial and temporal variability in the atmosphere. Detailed aerosol properties can only be measured in situ, and a few ground-based measurement networks for this purpose have been established. These include the Global Atmospheric Watch (GAW) aerosol program (<http://www.wmo.int/gaw/sag/aerosol>) and the global AERONET network of ground-based sun photometers (http://aeronet.gsfc.nasa.gov/new_web/system_descriptions_operation.html) as well as various regional networks, such as the European Monitoring and Evaluation Programme EMEP (<http://www.emep.int/>), EUSAAR (Philippin et al., 2009), and the US Interagency Monitoring of Protected Visual Environments IMPROVE (<http://vista.cira.colostate.edu/improve/>). Information on the



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vertical structure of aerosol properties can be obtained from aircraft, balloon and lidar measurements and from model simulations. Remote sensing with satellite instruments provide aerosol data over large spatial areas, but the information is limited to particles in the optically-active size range, i.e. particles larger than about 100 nm in diameter. Passive instruments on satellites are currently able to provide column-integrated aerosol properties, such as the aerosol optical depth (AOD) at several wavelengths, and space-borne lidars provide vertical profile information on a global scale. Some instruments provide also microphysical properties such as the fine and coarse mode fraction, effective radius, and information on particle shape or aerosol components (see e.g. Kokhanovsky and de Leeuw, 2009 and de Leeuw et al., 2011 for a comprehensive overview of satellite capabilities). Instruments with multiple viewing angles provide information on vertical structures in volcanic ash and fire plumes (e.g. Kahn et al., 2007, 2008; Muller et al., 2007).

A key phenomenon associated with the atmospheric aerosol system is the nucleation and subsequent growth of nucleated aerosol particles. Field measurements have demonstrated nucleation to be a frequent phenomenon in the continental boundary layer, as well as in the free troposphere (Kulmala and Kerminen, 2008, and references therein; Manninen et al., 2010). Direct observational evidence has been obtained that particles nucleated in the atmosphere are able to grow into cloud condensation nuclei (CCN) sizes (Lihavainen et al., 2003; Laaksonen et al., 2005; Wiedensohler et al., 2009) and participate in cloud droplet formation (Kerminen et al., 2005). Global model simulations suggest that nucleation is very likely the dominant source of particles in terms of their number concentration in the global atmosphere (Spracklen et al., 2006, 2010; Yu et al., 2010), and a significant contributor to global CCN concentrations (Spracklen et al., 2008; Merikanto et al., 2009; Pierce and Adams, 2009; Yu and Luo, 2009). As a result, nucleation has the potential to influence cloud properties and global radiative forcing (Wang and Penner, 2008; Makkonen et al., 2009; Merikanto et al., 2010; Kazil et al., 2010).

Combination of satellite data with either model simulations or in situ observations has been successfully used in several applications, including surface air quality predictions (e.g. Martin, 2008; Hoff and Christopher, 2009), evaluation of emission inventories (e.g. Lamsal et al., 2011; Lee et al., 2011), and constraining the radiative effects by aerosols (e.g. Myhre, 2009). Our understanding on atmospheric nucleation relies essentially on field and laboratory experiments, theoretical calculations and model studies (Kerminen et al., 2010), with practically no use of satellite data. Satellite measurements of nucleated particles are complicated by their relatively slow growth to optically active sizes in the atmosphere (e.g. Tunved et al., 2006). Therefore, alternative methods to trace these particles on regional and global scales using satellite data need to be explored.

In this paper we propose the use of proxies, i.e. parameterizations for the concentrations of nucleated particles in terms of satellite-observable quantities. These proxies are developed based on our best understanding on the atmospheric nucleation and growth processes. To the extent possible, the proxies are evaluated using detailed information from long-term ground-based measurements. The considered proxies describe the total number concentration of nucleation mode particles, that is, particles smaller than about 25–30 nm in diameter. Our emphasis is put on the continental boundary layer, since nucleation in that region is both more active and better understood than nucleation taking place over marine areas (Vuollekoski et al., 2009; Kazil et al., 2010; O'Dowd et al., 2010). In terms of the global aerosol number budget, free-troposphere nucleation is probably extremely important as well (e.g. Merikanto et al., 2009), but our approach is not suitable for tracing particles formed in that region due to long times scales associated with the life cycle of these particles.

2 Proxies for nucleation mode particle number concentrations

The balance equation for the particle concentration in the nucleation mode, N_{nuc} , can be written as

$$\frac{dN_{\text{nuc}}}{dt} = J - N_{\text{nuc}} \times \text{CoagS}, \quad (1)$$

where J is the nucleation rate and CoagS is the average coagulation sink for the nucleation mode (Kulmala et al., 2001). The nucleation rate can be connected to the gas-phase concentration of the nucleating vapour, C , via the following general relation:

$$J = K_{\text{nuc}} \times C^n. \quad (2)$$

Here K_{nuc} is the so-called nucleation coefficient and the exponent n is related to the nature of the nucleation mechanism (McMurry and Friedlander, 1979; Kulmala et al., 2006). The nucleation coefficient takes into account the effect of factors other than the vapour driving the nucleation rate, including the ambient temperature and relative humidity and the presence of impurities (trace gases, ions) that influence the stability of nucleating clusters. The vapors dictating the nucleation rate are expected to be of extremely low volatility, so their gas-phase concentration can be approximated by the relation

$$\frac{dC}{dt} = Q - C \times \text{CS}. \quad (3)$$

Here Q is the source rate of vapour C due to chemical reactions and CS is the condensation sink (Kulmala et al., 2001).

In the following we derive proxies for the nucleation mode particle number concentration, N_{nuc} , in two different situations of boundary-layer nucleation: (1) regional nucleation driven by photochemistry and occurring typically over spatial scales of hundreds of kilometers (Kulmala and Kerminen,

2008), and (2) primary nucleation that takes place in the immediate vicinity of highly localized sources, or at scales substantially smaller than those resolved by large-scale modeling frameworks (Luo and Yu, 2011). We also discuss how to apply these proxies when relying on satellite measurements.

2.1 Regional atmospheric nucleation

The times scales, over which the nucleation mode particle number concentration and nucleating vapour concentration reach a pseudo-steady state with respect to sources and sinks is equal to the inverse of CoagS and CS, respectively (Kerminen et al., 2004). In regional nucleation, the assumption of such a steady state is reasonable. By setting the left hand side of Eqs. (1) and (3) equal to zero, indicative of the pseudo-steady state, and by combining Eqs. (1) to (3), we obtain:

$$N_{\text{nuc}} = K_{\text{nuc}} \frac{Q^n}{\text{CS}^n \text{CoagS}}. \quad (4)$$

For nucleation mode particles having a mean diameter of d_{nuc} , the coagulation sink is proportional to the condensation sink via the following relation (Lehtinen et al., 2007):

$$\text{CoagS}(d_{\text{nuc}}) = \text{CS} \left(\frac{d_v}{d_{\text{nuc}}} \right)^m. \quad (5)$$

Here d_v is the diameter of the condensing vapor molecule, i.e. the vapor for which the value of CS is determined, and the exponent m varies between about 1.5 and 1.9 depending on the shape of the pre-existing particle number size distribution. The mean diameter of the nucleation mode, d_{nuc} , varies with time. By noting this and combining Eqs. (4) and (5), we then obtain:

$$N_{\text{nuc}} = K \frac{Q^n}{\text{CS}^{n+1}}. \quad (6)$$

The factor K now carries the information on both the nucleation coefficient and the size-dependent relation between CoagS and CS according to Eq. (5).

Atmospheric observations (Riipinen et al., 2007; Kuang et al., 2008; Paasonen et al., 2010), as well as recent laboratory measurements (Metzger et al., 2010; Sipilä et al., 2010), suggest that the nucleation rate scales to the power 1–2 of the nucleating vapor concentration. In most cases a clear correlation between J and C is obtained by assuming sulphuric acid to be the sole driver of the nucleation process, whereas in some cases an additional vapour, most likely an organic one, is needed (Paasonen et al., 2010). Gaseous sulphuric acid (SA) is produced by the oxidation of sulfur dioxide with the hydroxyl radical:

$$Q_{\text{SA}} = k_1 [\text{OH}][\text{SO}_2], \quad (7)$$

whereas condensing organic vapors can be produced by multiple oxidants (Kroll and Seinfeld, 2008). There are some

indications, however, that the organics most likely to participate in nucleation (NUCORG) are those formed preferably by the OH-initiated oxidation of organic precursor compounds, ORG (Hao et al., 2009):

$$Q_{\text{NUCORG}} = k_2 [\text{OH}][\text{ORG}]. \quad (8)$$

The gaseous OH concentration, rarely available from measurements, is usually directly proportional to the ultraviolet radiation intensity, UV, in the lower troposphere (Rohrer and Berresheim, 2006). The performance of UV as a proxy for the OH-initiated oxidation of SO_2 has been demonstrated by comparing predicted and measured gaseous sulphuric acid concentrations (Petäjä et al., 2009).

By setting the exponent n to either 1 or 2, by assuming that $[\text{OH}]$ is proportional to UV, and by combining Eqs. (6), (7) and (8), we obtain four potential proxies for N_{nuc} :

$$N_{n,1} = \frac{\text{UV} [\text{SO}_2]}{\text{CS}^2}, \quad (9)$$

$$N_{n,2} = \frac{\text{UV}^2 [\text{SO}_2]^2}{\text{CS}^3}, \quad (10)$$

$$N_{n,3} = \frac{\text{UV} [\text{ORG}]}{\text{CS}^2}, \quad (11)$$

$$N_{n,4} = \frac{\text{UV}^2 [\text{ORG}]^2}{\text{CS}^3}. \quad (12)$$

In analyzing field measurements we may apply the proxies (9) and (10) as such, whereas proxies (11) and (12) require estimation of either the organic precursor vapor concentration, $[\text{ORG}]$, or the concentration of organic vapors participating in nucleation and very initial growth of the nuclei, $[\text{NUCORG}]$. If the latter quantity is used, the proxies (11) and (12) will be reduced to the following forms: $N_{n,3} = [\text{NUCORG}]/\text{CS}$, $N_{n,4} = [\text{NUCORG}]^2/\text{CS}$. The value of $[\text{NUCORG}]$ can be derived from measured nuclei growth rates within a factor of about 2 (Paasonen et al., 2010).

2.2 Primary nucleation

The steady-state assumptions made in the previous subsection are no longer valid for primary nucleation because the nucleation process can be extremely rapid, as it is in case of vehicular emissions, or because the nucleating air parcels are diluted very rapidly (Kerminen et al., 2004). In urban areas with primary nucleation resulting from traffic exhaust, concentrations of nucleation mode particles have been observed to correlate with nitrogen dioxide (NO_2) concentrations (e.g. Fernandez-Camacho, 2010). When spread over larger volumes of ambient air, primary nuclei are expected to be scavenged by pre-existing larger particles according to Eq. (1). By assuming a steady state for this process we obtain

$$N_{n,5} = \frac{[\text{NO}_2]}{\text{CS}}. \quad (13)$$

This is a potential proxy for primary nucleated particles resulting from traffic emissions, or any other combustion sources that both emit significant amounts of nitrogen oxides and initiate primary nucleation in the atmosphere.

In large-scale models, a fraction of the sulfur emissions related to anthropogenic combustion sources is assumed to be in the form of primary particles and a fraction of these particles are often assumed to be nucleation mode particles resulting from sub-grid scale nucleation (see Luo and Yu, 2011, and references therein). By applying the same reasoning as above, the following satellite proxy for these nuclei can be derived:

$$N_{n,6} = \frac{[\text{SO}_2]}{\text{CS}}. \quad (14)$$

This is a potential proxy primary nuclei associated with strong SO_2 emitters such as some coal-fired power plants and smelters.

2.3 Satellite applications

In case of satellite measurements, the proxies given by Eqs. (9) to (14) need to be simplified further. To start with, we need to replace CS with a proper column-integrated quantity. Here we propose the aerosol optical depth (AOD) for this purpose for several reasons. First, the satellite-derived AOD has been successfully used to trace surface particulate matter concentrations in air quality applications (Hoff and Christopher, 2009), in addition to which it has turned out to be a good tracer for atmospheric cloud condensation nuclei concentrations (Andreae, 2009). Second, both vapor condensation and light scattering are roughly proportional to the aerosol surface area distribution. The relatively good correlation between CS and aerosol light scattering coefficient has been confirmed by field measurements (Virkkula et al., 2011). Third, due to their similar dependence on the particle size, CS and AOD are expected to respond to changes in the ambient relative humidity in a similar manner. The apparent drawback with our approach is that as a column-integrated property AOD is unable to take into account the influence of mixed-layer height on CS, nor the effects of elevated aerosol layers on the relation between CS and AOD. The performance of replacing CS with AOD will be investigated in Sect. 3.1.

Observation of sulfur dioxide by satellites is extremely challenging and usually limited to strongly polluted regions and major plumes originating from power plants, smelters or volcanic eruptions (see Veefkind et al., 2011, and references therein). Compared with SO_2 , satellite measurements of NO_2 column burdens typically encountered in the lower troposphere are much more accurate. The connection between NO_2 and AOD, as retrieved from satellites, has been shown to reflect different aerosol source types to be consistent with the corresponding connection obtained from global model simulations (Veefkind et al., 2011).

Few organic compounds can be detected with satellite instruments, and the only one having a clear association with organic aerosol precursors is formaldehyde (HCHO). Column retrievals of HCHO have been successfully used to constrain non-methane hydrocarbon emissions from biogenic and biomass burning sources (Stavrakou et al., 2009) and, in some cases, to trace secondary organic aerosol concentrations (Veefkind et al., 2011). These findings suggest that it might be possible to derive [ORG] in the proxies given by Eqs. (11) and (12) using satellite data on HCHO. Before doing that, however, the potential connection between HCHO and ORG should be investigated by in situ field measurements.

The above discussion points out that neither $[\text{SO}_2]$ nor [ORG] are usually available from satellite measurements. One, yet by no means ideal, solution for this problem is to remove these two quantities from the proxies given by Eqs. (9) to (12) by setting them constant, which is equal to assuming that it is photochemistry rather than the exact concentration of any trace gas that dictates the nucleation rate. We will discuss the consequences of this very crude approximation in Sects. 3.1 and 3.2.

3 Evaluation and preliminary results

In this section, we evaluate selected proxies against in situ field measurements and then apply them on the global scale using satellite retrievals. The main purpose of the evaluation, conducted in Sect. 3.1, is to find out how critical it is to have knowledge on the SO_2 concentration when applying the proxy given by Eq. (9), and whether replacing CS with AOD can be considered reasonable. In Sect. 3.2 we investigate the potential of using satellite-derived column SO_2 and NO_2 concentrations in association with our proxies and, most importantly, discuss the overall performance of the proxy given Eq. (9) in the global atmosphere after setting the SO_2 concentration constant and replacing CS with AOD.

3.1 Evaluation of selected proxies against in situ measurements

The ground-based data used in this work were obtained from measurements at the SMEAR II station in Hyytiälä, Southern Finland, located in the boreal forest (Hari and Kulmala, 2005). Size distributions of 3–1000 nm particles have been measured at the SMEAR II station continuously since 1996 (e.g. Kulmala et al., 2010). From these data we obtain both the nucleation mode particle number concentration, as well as the condensation sink (see Table 1). Here we take nucleation mode particles to be all particles smaller than 25 nm in mobility diameter. Condensation sink is calculated from the number size distributions according to the method presented in Kulmala et al. (2001) accounting for particle hygroscopic growth according to the parameterization by Laakso

Table 1. Mean, median and percentiles (10th and 90th) for the measured data set used in the comparison of proxy performance in Hyytiälä. Measured nucleation mode particle concentrations N_{nuc} were compared to Eq. (9) (proxy utilizing UVB, SO_2 , CS and AOD). The comparisons are shown in Figs. 1 and 2. In the comparisons data was averaged to 30 min time resolution.

| | mean | median | 10th percentile | 90th percentile |
|---|------|--------|-----------------|-----------------|
| $N_{\text{nuc}} \text{ (cm}^{-3}\text{)}$ | 1100 | 320 | 53 | 2730 |
| UVB (W m^{-2}) | 0.45 | 0.63 | 0.20 | 2.02 |
| SO_2 (ppb) | 0.18 | 0.10 | 0.03 | 0.36 |
| CS (10^{-3} s^{-1}) | 3.4 | 2.7 | 1.0 | 5.9 |
| AOD | 0.10 | 0.09 | 0.05 | 0.18 |

et al. (2004). SO_2 concentration has been measured continuously since 1996 by a fluorescence analyser (TEI 43 BS, Thermo Environmental, Franklin, MA, USA). The detection limit of the instrument is 0.1 ppb.

Aerosol optical depth measurements were started at the SMEAR II station in February 2008. They are part of the global AERONET network of ground-based sun photometers (Holben et al., 1998). AOD is measured at 8 different wavelengths, and here we use the wavelength of 500 nm as this is closest to the satellite-based observations used in the global proxies. The sequence of measurements, from which AOD and aerosol microphysical parameters are derived, is described at the AERONET webpage (http://aeronet.gsfc.nasa.gov/new_web/system_descriptions_operation.html). Sun photometer observations are available only when the air mass, i.e. the optical path length through the atmosphere, is equal to 7 or less. Due to the northern location of Hyytiälä, this excludes the AOD data availability from about mid-November to mid-February.

Figure 1 (left panel) shows the comparison between measured nucleation mode particle number concentrations and those derived from the proxy given by Eq. (9) based on 22 months of in situ measurements during 2008–2010 at the SMEAR II station. The two quantities are positively correlated ($r = 0.54$), but there is also lot of scatter in the data points. The main reason for the scatter in this figure is likely that the proxy assumes a very simple dependence of the nucleation rate, J , on trace gas concentrations and environmental conditions (nucleating vapour is sulphuric acid; K_{nuc} is constant and $n = 1$ in Eq. 2). In reality, the value of n has been found to be somewhere between 1 and 2 at SMEAR II and elsewhere, whereas the value of K_{nuc} may vary up to an order of magnitude between individual nucleation events at any particular site (Riipinen et al., 2007; Kuang et al., 2008; Paasonen et al., 2010). Another reason for the scatter is the relatively strong dependence of the nuclei removal rate by coagulation on the nuclei size (Eq. 5). This makes the life

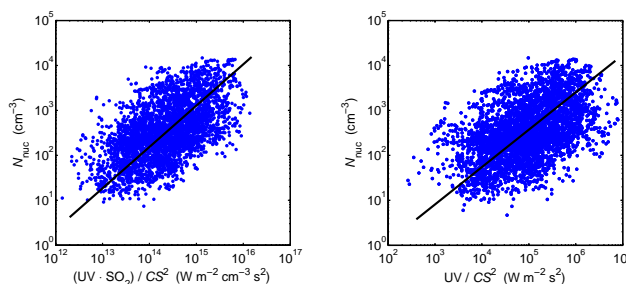


Fig. 1. Measured number concentration of nucleation mode particles (N_{nuc} , diameter 3–25 nm) as a function of the proxy given by Eq. (9) (left) and the same proxy with SO_2 concentration left out (right). Each data point represents 30 min averaged data. The black lines show linear least-squares fit to the data points. Left: $\log(y) = 0.92 \times \log(x) - 10.69$ ($r = 0.54$, $p < 0.001$); Right: $\log(y) = 0.83 \times \log(x) - 1.58$ ($r = 0.49$, $p < 0.001$).

time and thereby the concentration of nucleation mode particles sensitive to their growth rate. Nuclei growth rates have been observed to vary by a factor of about 2–5 at individual measurement sites (e.g. Manninen et al., 2010).

If we neglect the influence of SO_2 concentration variations on our proxy, i.e. when we set $[\text{SO}_2]$ to be constant in Eq. (9), the correlation between the measured and proxy-derived nucleation mode particle number concentration decreases only slightly from 0.54 to 0.49 (Fig. 1, right panel). The relatively moderate influence of SO_2 on the performance of the proxy might appear surprising, given the strong association between the nucleation rate and gaseous sulphuric acid concentration observed at the SMEAR II station (Riipinen et al., 2007; Nieminen et al., 2009). On the other hand, this finding reflects the complexity by which the nucleation mode particle number concentration depends on the combination of the whole photochemistry (UV radiation intensity) and sinks for both nucleated particles and their precursor vapours (CS). Although knowing the exact magnitude of the SO_2 concentration appears not to be crucial at the SMEAR II station, we would like to stress here that the situation may be totally different in regions having either exceptionally high or very low SO_2 concentration levels.

Perhaps the most crucial of our assumptions is to replace CS with AOD. Figure 2 demonstrates how this replacement affects the performance of our proxy in case of our in situ measurements. The correlation between the measured and proxy-derived nucleation mode particle number concentrations reduces now down to 0.25 when the SO_2 concentration is taken into account and to 0.23 when it is not. We may conclude that while our solution to replace CS with AOD is necessary in order to apply the proxies to a global scale using satellite data, it is clearly not the ideal one.

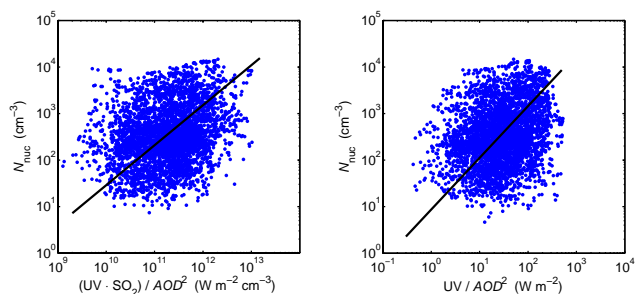


Fig. 2. Same as Fig. 1, but with CS replaced with AOD. Left: $\log(y) = 0.86 \times \log(x) - 7.141$ ($r = 0.25$, $p < 0.001$); Right: $\log(y) = 1.11 \times \log(x) + 0.94$ ($r = 0.23$, $p < 0.001$).

3.2 Preliminary predictions for the global troposphere

SO_2 and NO_2 concentrations are available from satellite-based spectrometers such as SCIAMACHY, GOME-2, TOMS, and OMI. The Total Ozone Mapping Spectrometers (TOMS) onboard several satellite platforms followed up by Ozone Monitoring Instruments (OMI) have provided a UV data record of more than 30 yr. In our analysis we use monthly data of OMI UV irradiance at 310 nm from the year 2006. The primary aerosol products from the MODIS instruments aboard the Terra and Aqua satellites are the AOD and the fine aerosol weighting (FW) at a wavelength of 550 nm. The spatial resolution is $10 \times 10 \text{ km}^2$. In our analysis we used the monthly mean satellite data available from the GIOVANNI website (<http://disc.sci.gsfc.nasa.gov/>). Both the MODIS AOD (e.g. Levy et al., 2010) and the OMI UV (e.g. Tanskanen et al., 2007) products have been extensively validated against ground-based measurements.

To begin with, we investigate whether column SO_2 retrievals could be used with our proxies despite the detection limit for this compound by satellites. For this purpose we calculated three-month average SO_2 concentration fields from the satellite data. The concentrations were close to zero, including also negative values, and without any clear and justified large-scale geographical patterns (Fig. 3). Therefore, in our subsequent proxy analysis only satellite-derived UV and AOD data are used.

Figure 4 shows the proxy constructed according to Eq. (9) with the two main assumptions considered in the previous section, i.e. CS replaced with AOD and $[\text{SO}_2]$ assumed to be constant. We do not include the influence of SO_2 variations for the reasons explained above. Four different seasons are shown: December–February (DJF), March–May (MAM), June–August (JJA), and September–November (SON). Our proxy predicts that the Southern Hemisphere is the dominating source for nucleation mode aerosol particles during DJF and MAM, whereas the Northern Hemisphere dominates during JJA. During SON, active regions for nucleation are predicted for both hemispheres. The relative importance of AOD and UV radiation in causing the seasonal cycle of the

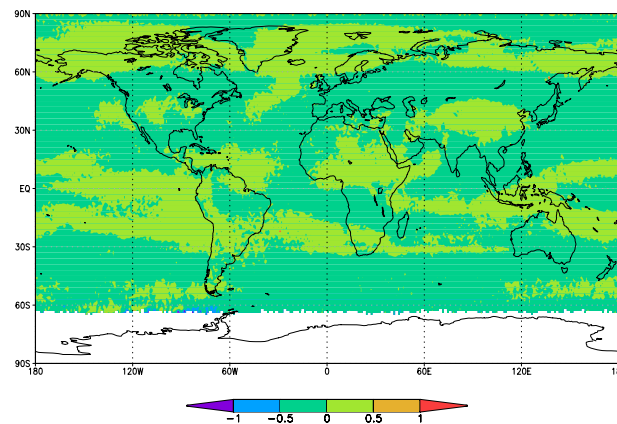


Fig. 3. SO_2 Column Amount (Middle Troposphere) from OMI L2G product in Dobson Units for June, July and August in 2006. The color scale has been restricted; the lowest and highest spots are close to -2 and 2 , respectively.

proxy varies geographically depending on the latitude, which determines the variability of the UV radiation intensity, and how the main aerosol source and sinks behave annually.

Many of the patterns shown in Fig. 4 are qualitatively consistent with field observations and our current understanding on atmospheric nucleation (e.g. Kulmala et al., 2004; Kerminen et al., 2010). These include the extremely frequent and strong nucleation taking place over South Africa throughout the year (Vakkari et al., 2011), active yet less intensive nucleation observed over the boreal forest areas during the summer part of the Northern Hemisphere (Tunved et al., 2006; Dal Maso et al., 2008), and frequent nucleation taking place in the South-East Australian rainforest during most parts of the year (Sun et al., 2008). Field measurements show relatively frequent nucleation taking place over many parts of Central and Southern Europe almost throughout the year (Jaatinen et al., 2009; Manninen et al., 2010), over the North-Eastern United States outside the winter period (Stanier et al., 2004; Qian et al., 2007; Pryor et al., 2010), as well as in Beijing, China (Wu et al., 2007). Our proxy captures some of these features but totally fails in case of China. A possible reason for this is the very high SO_2 concentration, and thereby very active role of it, in nucleation taking place over polluted regions of China. Finally, our proxy predicts very high nucleation mode particle number concentrations over large areas in Eastern South America and in some parts of the Amazon Basin. While there are practically no field data to confirm whether the pattern predicted for Eastern South America is correct or not, measurements conducted in the Amazon Basin show little evidence for near-surface regional-scale nucleation (Ahlm et al., 2009; Martin et al., 2010). The discrepancies between the proxy predictions and existing observations in the Amazon Basin, if real, might be related to too low SO_2 concentrations to initiate nucleation there (Martin et al., 2010). Such a feature is not captured by a proxy assuming

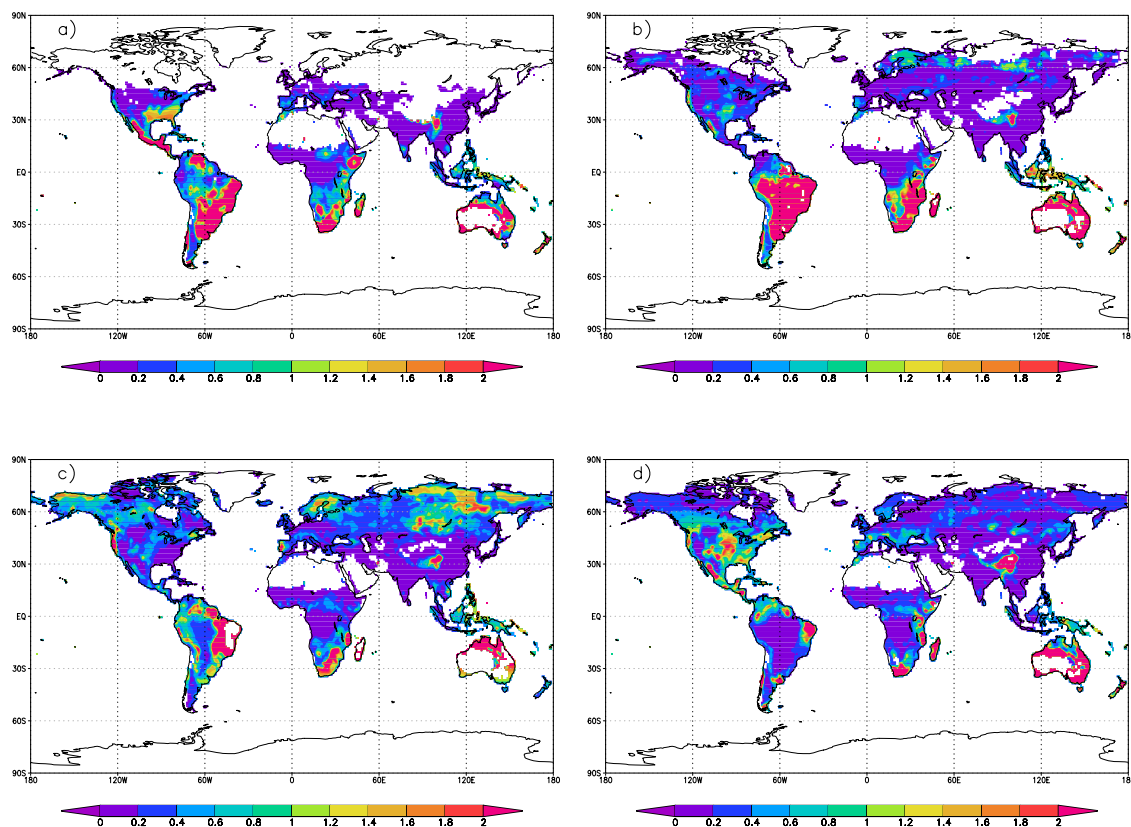


Fig. 4. The global distribution of the proxy given by Eq. (9), after replacing CS with AOD and assuming SO_2 concentration to be constant, for the different seasons of the year 2006: DJF (a), MAM (b), JJA (c), and SON (d). The unit of UV is $\text{mW m}^{-2} \text{nm}^{-1}$, AOD is unitless. Moreover, UV/AOD^2 is normalized by a factor of 10 000 and the color scale is restricted to between 0 and 2, in order to better show the geographical patterns.

a constant SO_2 concentration. Besides field measurements, Fig. 4 shows many similarities to global model simulations made with a nucleation mechanism that is consistent with the proxy given by Eq. (9) (Spracklen et al., 2006).

Figure 5 shows an example of the proxy for primary nucleated particles (Eq. 13). The most significant concentrations are predicted over the polluted regions in South Africa, which would add to active regional nucleation predicted for this region. This is consistent with observations (Vakkari et al., 2011), even though the relative importance of regional and primary nucleation may be difficult to separate from field measurements. Our proxy does not predict any primary nucleation over the polluted regions of China which, again, might be due to the important role of SO_2 in driving nucleation there.

4 Concluding remarks

We have derived proxies based on physical processes to estimate the concentration of nucleation mode particles. The proxies given by Eqs. (9) to (12) describe nucleation mode particle concentrations resulting from regional-scale

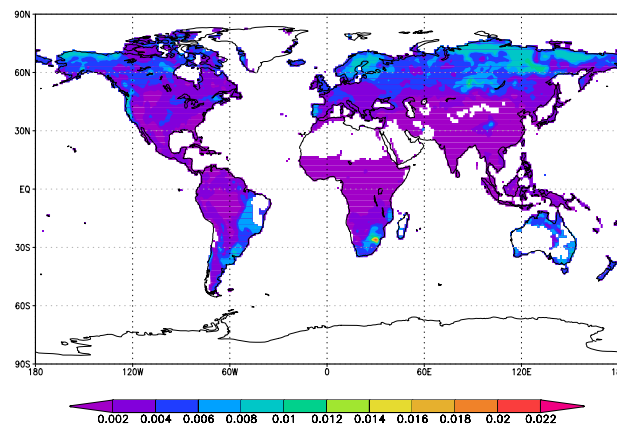


Fig. 5. The proxy given by Eq. (13) for the JJA season of the year 2006. The unit of NO_2 is $10^{15} \text{ molec cm}^{-2}$, AOD is unitless. Moreover, NO_2/AOD is normalized by a factor of 10 000.

atmospheric new particle formation, whereas the proxies given by Eqs. (13) and (14) describe the contribution of direct emissions of nucleation mode particles (primary nuclei). When applied to satellite data, further simplifications in proxies are currently needed, such as replacing the condensation sink (CS) with the aerosol optical depth (AOD) and neglecting the influence of sulfur dioxide (SO₂) concentration variations or organic compounds on the nucleation rate.

The global pattern of nucleation mode particle number concentration predicted by satellite data had similarities to both observations and global model simulations, but revealed also several problems associated with our current approach. Problems were evident in regions where the nucleation rate is apparently sensitive to the SO₂ concentration level, such as the polluted areas of China and regions with very low SO₂ concentrations. Other problematic locations seem to be those having a significant contribution from primary nuclei associated with either NO₂ or SO₂ emissions. Furthermore, our current proxies cannot properly take into account the influence of organic compounds on nucleation and subsequent particle growth.

In order to be able to improve our results, more sophisticated products from satellite data are clearly needed. The present products like column NO₂ concentration, and even more so the column SO₂ concentration, are not good enough to distinguish any details in non- or less-polluted conditions. The satellite proxies would definitely benefit from having an explicit relation between CS and AOD that would take into account the influence of mixed layer height, the relative role of coarse and fine particles on CS and AOD, and the presence of potential aerosol layers aloft. More work is clearly needed in which satellite retrieval of organic compounds is combined with new information from in situ field experiments. The use of satellite products such as the UV actinic flux would be better than the surface irradiance used here. Currently such a data product is not available.

One should also explore the possibility to combine satellite data with information obtained from global or regional model simulations, as has been done in some other applications (see Sect. 1). An evident example in this regard would be to use some sort of time-averaged SO₂ concentration field predicted by a global chemical transport model when applying the proxies 9 or 10 in different regions and at different times of the year. Model simulations might also be helpful in searching for a useful linkage between CS and AOD, or in attempting to include the influence of organic compounds on proxy predictions.

With our proxy method we have been able to significantly contribute to the application of satellite data to obtain information on aerosol dynamics in the global atmosphere. In future when this approach will be developed further and evaluated properly against long-term field observations, this knowledge might be utilized in global climate studies and in identifying new regions where continuous ground-based measurements should be started.

Acknowledgements. This research has received funding from ERC Advanced Grant “ATMNUCLE” no. 227463, the Academy of Finland Center of Excellence program (project no. 1118615), and the ESA Contract no. 4200023053/10/I-LG, STSE-ALANIS-Atmosphere-Land Interactions Study Theme 3: Aerosols.

Edited by: A. Laaksonen

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