



# New particle formation events arising from painting materials in an indoor microenvironment



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## HIGHLIGHTS

- New particle formation events were observed arising from painting materials indoors.
- Nucleation rates were measured in conjunction with high TVOC concentrations.
- High human exposure levels of particle number concentration were observed.

## ARTICLE INFO

### Article history:

Received 10 April 2014

Received in revised form

20 November 2014

Accepted 21 November 2014

Available online 22 November 2014

### Keywords:

Aerosol growth

New particle formation

Indoor particle emissions

Paint emissions

## ABSTRACT

Particulate matter (PM) number size distribution and mass concentration along with total volatile organic compounds (TVOC) were measured during emissions from painting materials inside an indoor microenvironment. The emission sources were derived from oil painting medium and turpentine used for painting. Two sets of measurements (10 experiments) were conducted in a laboratory room of 54 m<sup>3</sup>. New particle formation events were observed in all 10 experiments. The nucleation events lasted on average less than one hour with an average growth rate  $33.9 \pm 9.1$  nm/h and average formation rate  $21.1 \pm 8.7$  cm<sup>-3</sup>s<sup>-1</sup>. After the end of the nucleation event, a condensational growth of indoor particles followed with average growth rate  $11.6 \pm 2.8$  nm/h and duration between 1.4 and 4.1 h. High concentrations up to 3.24 ppm were measured for the indoor TVOC concentrations during the experiments. Simultaneous mass and number size concentration measurements were performed outdoors where no new particle formation event was observed. It is the first time that high nucleation rates indoors were observed in conjunction with high TVOC concentrations originating from painting materials which resulted to high exposure concentration levels of particle number concentration.

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## 1. Introduction

New particle formation events and condensational growth of particles in the outdoor air were investigated extensively the last decades (Kulmala et al., 2004; Holmes, 2007). Nucleation events were observed in many different sites in the atmospheric air including the free troposphere, lower stratosphere, in arctic, in forest, in coastal areas and in cities (Kulmala et al., 2004; Curtius, 2006; Holmes, 2007).

Several mechanisms are proposed for the occurrence of new particle formation, such as the binary homogenous nucleation of sulphuric acid–water system, the ternary homogenous nucleation

of sulphuric acid–water–ammonia system, the heterogeneous nucleation on pre-existing particles and the heterogeneous ion-induced nucleation (Kulmala et al., 1991; Lazaridis, 2001; Housiadas et al., 2004; Seinfeld and Pandis, 2006).

A nucleation event provokes the generation of new particles at low particle sizes, usually of few nanometers. Particles in the nucleation mode (<0.1 μm) are the most harmful to human health due to their ability to penetrate very easily through the human respiratory tract. Several health effects (cardiovascular, respiratory, lung cancer) are connected with the presence of ultrafine particles to the ambient air (Pope and Dockery, 2006).

However, ultrafine particles emitted to the indoor air originate from primary sources as well. Almost every human activity generates particles in the indoor air. The particle characteristics of each emission activity are closely related to the primary source. Usual indoor sources due to human activities is combustion, cooking,

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cleaning, smoking, printing and walking (Ferro et al., 2004; He et al., 2004; Hussein et al., 2006; Wallace, 2006; Géhin et al., 2008; Glytsos et al., 2010; Wang et al., 2012; Semple et al., 2012). High emission rates or concentrations are reported in the previous studies with substantial increase of indoor ultrafine particles during the described activities.

On the other hand, indoor emissions of chemical substances from building materials, household products and furnishings may lead to secondary organic aerosol formation (Weschler, 2001; Singer et al., 2006; Aoki and Tanabe, 2007; Uhde and Salthammer, 2007; Weschler, 2009). Semivolatile compounds found in indoor environments are provoked by several sources (Weschler and Nazaroff, 2008). In particular, painting materials are a very common source and it involves a wide range of volatile organic compounds (VOC) (Guo et al., 1998; Weschler, 2009). Emissions from wet materials like paints are dominated by evaporation at the beginning and by internal diffusion afterwards (Yang et al., 2001; Zhang and Niu, 2003). A previous study reported formation of ultrafine particles during the reaction of VOCs and ozone (Fan et al., 2005), suggesting that VOCs may behave as precursor material. Moreover, ultrafine particle formation was also observed in a study of emissions from natural paint in presence of ozone underlying that the paint may play a significant role in generation of new particle in indoor environments (Lamorena et al., 2007).

New particle formation events indoors, associated with the presence of VOCs and ozone, conducted in real environments are already reported in the scientific literature (Vartiainen et al., 2006; Betha et al., 2011; Hovorka and Branis, 2011; Quang et al., 2013). In the work by Quang et al. (2013) nucleation events observed inside a big office building. However, the observed nucleation was the result of outdoor particles transported indoors due to high air exchange rate of the building with the outdoor air. Vartiainen et al. (2006) investigated the formation of indoor particles as a result of  $\alpha$ -limonene oxidation. Betha et al. (2011) suggested that the formation of ultrafine particles caused possibly by the ozone-induced oxidation of VOCs, whereas, Hovorka and Branis (2011) associated the nucleation events into a constructing hall with emissions from the paint spraying. Volatile organic compounds emitted from the paint were more likely to cause ultrafine particle formation event rather than the penetration from outdoors due to low air exchange rate and high residence time of the ultrafine particles inside the hall, as suggested by the authors.

This study presents an analysis of new particle formation events from painting materials that took place in a laboratory room. The objectives were to measure particle number and mass concentrations emitted from painting materials, to investigate the aerosol particle number size distribution during the emission process and further to examine the formation rates and condensational growth of the particles during the new particle formation events.

## 2. Materials and methods

### 2.1. Experimental setup

The measurements were performed from September 2013 to January 2014 in the atmospheric aerosol laboratory at the Technical University of Crete. The area of the laboratory is 19 m<sup>2</sup> of rectangle shape and its volume is 54 m<sup>3</sup> with one door and one window placed at the opposite side. During the measurements, the instruments were placed on the bench next to each other. At the first two experiments, two persons were present in the laboratory during the emission period and only one person was present in the laboratory during the rest of the experiments. The window and the door were closed at all times. Indoor temperature and humidity were continuously recorded in a nearby laboratory room (of the

same dimensions) using the indoor sensors of a Davis Vantage PRO 2 meteorological station. Previous comparative measurements in the two laboratory rooms showed that there were no significant differences in the indoor conditions and therefore the data from the nearby room can be used, since during the nucleation experiments no air-condition or heating devices were used. Average indoor temperature ranged from 19.6 °C to 28.5 °C while indoor humidity ranged from 42% to 55% depending on the time of the year that the experiments were conducted. Furthermore, the air exchange rate between the laboratory room and the outdoor environment was measured using CO<sub>2</sub> as a trace gas and the average value for all the experimental periods was 0.16 h<sup>-1</sup>.

The painting materials that were used consisted of water mixable oil medium (Artisan Series, Winsor & Newton) (linseed oil) and turpentine medium (Winsor & Newton) (composed mainly from monoterpenes alpha-pinene and beta-pinene). Oil colours (Old Holland) were used during the first two experiments for painting on linen canvas.

The experiments were performed in two periods, each consisting of 5 experiments (10 experiments in total). The measurements were conducted in 3 phases characterised as follows:

<u>Phase 1:</u>	Background measurement:	0–60 min	(Empty laboratory)
<u>Phase 2:</u>	Emission period:	60–150 min	(Painting and/or opened bottles)
<u>Phase 3:</u>	Removal period:	150–330 min	(Empty laboratory)

During phases 1 and 3 no person was present in the laboratory, whereas, during phase 2 only one person was present. Only on the experiments conducted on 26/09/13 and 02/10/13, two persons were present during phase 2. In these two experiments, the activity period (phase 2) consisted of painting a picture on a canvas with painting oil colours with a simultaneous opening of the medium and turpentine oils bottles. The emission period in the other experiments consisted of opening 10 bottles with oil medium and turpentine, which, were brought in the laboratory just before the start of phase 2 (emission period). The bottles with turpentine oils were placed close to the instrumentation (distance of 0.5 m) for a time period of 90 min. After the emission period, the bottles were closed and removed from the laboratory room. The door and window were closed during all three phases.

### 2.2. Instrumentation

The indoor particle mass concentration was measured with a Dust Trak (Aerosol Monitor, Model 8520, TSI) and a Dust Trak II (Aerosol Monitor, Model 8532, TSI) instrument for a selected size of 10 μm particles (PM<sub>10</sub>). The Dust Trak and Dust Trak II instruments have a particle size range 0.1–10 μm. Dust Trak's flow rate was equal to 1.7 lpm, whereas, the Dust Trak II airflow rate was equal to 3.0 lpm. The log interval was set up for 1 min.

The indoor particle number concentration was measured with a P-Trak (Ultrafine Particle Counter, Model 8525, TSI), and a NanoScan SMPS (Model 3910, TSI) instrument. The P-Trak has a size range between 0.02 and 1 μm at a sample air flow rate 0.1 lpm. The portable NanoScan 3910 measuring principle is based on a unipolar charging of the particles, where, the particles are positively charged in a mixing chamber and sent to radial DMA (RDMA) for size classification and then are counted in isopropanol-based CPC. The NanoScan measures particle concentrations from 10<sup>2</sup> up to 10<sup>6</sup> particles/cm<sup>3</sup> with size resolution into 13 pre-setup channels in the size range of approximately 10–420 nm at a scan time of 60 s.

The outdoor particle number size distribution for the first measurement's period was measured with a SMPS+C (CPC Model

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